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October 25-28, 1988. Volume 1. Sessions 1, 2, 3, and 4

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B. B. Emmel, Compiler

Radian Corporation

P. O. Box 1300

Research Triangle Park, NC 27709

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Project Officers:

Charles B. Sedman

U. S. Environmental Protection  
Agency

Air and Energy Engineering  
Research Laboratory  
Research Triangle Park, NC 27711

P. T. Radcliffe

Electric Power Research Institute  
Desulfurization Processes Program  
Coal Combustion Systems Division  
3412 Hillview Avenue  
Palo Alto, CA 94304

AIR AND ENERGY ENGINEERING RESEARCH LABORATORY  
OFFICE OF RESEARCH AND DEVELOPMENT  
U. S. ENVIRONMENTAL PROTECTION AGENCY  
RESEARCH TRIANGLE PARK, NC 27711

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## PREFACE

The First Combined FGD and Dry SO<sub>2</sub> Control Symposium was held in St. Louis, Missouri, October 25 - 28, 1988. This Symposium, jointly sponsored by EPRI and EPA, had as its objective the exchange of technical and regulatory information on sulfur oxide control technology, including wet and dry scrubbers, emerging processes, and international developments in clean coal/acid rain technologies.

Eighty-five papers were presented by EPA and EPRI staff members, representatives of utility companies, equipment manufacturers, research and development companies, and university researchers. Specific topics discussed in symposium presentations included: retrofit economics, spray dryer technology, furnace sorbent injection topics (demonstration results, byproduct utilization, enhancements), wet FGD operation, municipal solid waste facilities, and post-combustion dry technologies. Also included were concurrent sessions on special topics of interest, including dry FGD, new technologies, and FGD improvement.

The Proceedings of this Symposium have been compiled in three volumes. Volume 1 contains papers from the following sessions:

- Session 1: International Overview
- Session 2: Retrofit Economics
- Session 3: Spray Dryer Technology
- Sessions 4A/4B: Furnace Sorbent Injection - Demonstrations/  
Integration/Byproduct Utilization.

Contained in Volume 2 are papers presented in these sessions:

- Sessions 5A/5B: FSI Impacts/Enhancements/Wet FGD Operation
- Session 6: Municipal Solid Waste Facilities

Contained in Volume 3 are papers presented in these sessions:

- Sessions 7A/7B/7C: Special Topics: Dry FGD Fundamentals/New Technologies/FGD Improvement
- Session 8: Post-Combustion Dry Technologies

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#### NOTICE

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## SESSION 1: INTERNATIONAL OVERVIEW

Chairman, James H. Abbott, EPA/AEERL

### FGD RESEARCH AND DEVELOPMENT STATUS IN THE U.S.

by: R. Dean Delleney  
James C. Dickerman  
Radian Corporation  
Austin, Texas 78759

#### ABSTRACT

A significant number of flue gas desulfurization (FGD) programs are sponsored by the United States Environmental Protection Agency (EPA) and the Electric Power Research Institute (EPRI) as indicated by their cooperative efforts in conducting this current and past symposia. Many of the papers presented here in St. Louis describe and report the results of projects funded by EPA and EPRI. This work and previous projects have been of substantial benefit to the electric utility industry in the United States (U.S.) and in foreign countries. However, other groups in the U.S. are also making substantial contributions to the advancement of FGD technology. The U.S. Department of Energy and several of the major coal-producing states are supporting a variety of work in the FGD area. The purpose of this paper is to summarize this work so that those with an interest in FGD will be better informed as to the spectrum of work being conducted in the U.S.

## INTRODUCTION

A significant number of flue gas desulfurization (FGD) programs are sponsored by the United States Environmental Protection Agency (EPA) and the Electric Power Research Institute (EPRI) as indicated by their cooperative efforts in conducting this current and past symposia. Many of the papers presented here in St. Louis describe and report the results of projects funded by EPA and EPRI. This work and previous projects have been of substantial benefit to the electric utility industry in the United States (U.S.) and in foreign countries. However, other groups in the U.S. are also making substantial contributions to the advancement of FGD technology. The U.S. Department of Energy and several of the major coal-producing states are supporting a variety of work in the FGD area. The purpose of this paper is to summarize this work so that those with an interest in FGD will be better informed as to the spectrum of work being conducted in the U.S.

The U.S. Department of Energy (DOE) has focused on advanced regenerable and in-duct sorbent injection technologies for post-combustion desulfurization of high-sulfur coal and combined  $\text{SO}_2/\text{NO}_x$  control processes. Additional work that DOE has cosponsored includes the High-Sulfur Test Center and full-scale demonstrations of furnace sorbent injection technology. In addition, under Round 2 of the Federal Clean Coal Technology (CCT II) program, DOE has selected 16 projects from the 54 proposals submitted. Seven of these projects which involve reduction of sulfur dioxide emissions from coal-fired utility boilers will be summarized in this paper. The advanced regenerable and combined  $\text{SO}_2/\text{NO}_x$  work will also be briefly discussed, but the in-duct scrubbing technologies are omitted because DOE is presenting a paper on this work in Session 8, Post-Combustion Dry Technologies, of this symposium. The full-scale furnace sorbent injection demonstrations are also discussed in other papers to be presented at this symposium and will not be included in this paper.

Two of the major high-sulfur coal-producing and consuming states are Illinois and Ohio. Both of these states have active state departments or offices working to expand the use of their coals in an environmentally sound and economically acceptable manner. One of the primary missions of these state departments is to promote the development and installation of clean coal technologies that permit the expanded use of high-sulfur Illinois and Ohio coal. In Illinois, the Department of Energy and Natural Resources (ENR) carries out this mission, while the Ohio Coal Development Office (OCDO) is the chief advocate for Ohio coal. One of the seven CCT II projects mentioned above is cosponsored by the Illinois ENR and two are cosponsored by the Ohio OCDO. Illinois is also a cosponsor of the in-duct sorbent injection pilot work to be performed at Central Illinois Public Service Company's Meredosia Station, and Ohio is cosponsoring the furnace sorbent injection demonstration at Ohio Edison's Edgewater Station. Other eastern coal-producing states are beginning to develop programs to promote use of their coal such as regulatory incentives for clean coal projects, but these programs are not as large or as active as those of Illinois and Ohio at this time.

### DEPARTMENT OF ENERGY CLEAN COAL TECHNOLOGY PROGRAM

The expansion of President Reagan's Clean Coal Technology Program provides \$536 million in Federal funds for advanced demonstration projects that employ as yet uncommercialized technologies for reducing air pollution from coal use. The CCT II Project Opportunity Notice attracted 54 proposals valued at more than \$5.3 billion to demonstrate a new generation of pollution control and power generating options for the U.S. coal industry. The objective of the CCT II projects is to

provide a technological approach to resolving concerns over acid rain while building a greater market for U.S. coal.

Seven of the projects selected under CCT II involve wet or dry FGD technologies. The sponsors, project title, and location for these seven projects are identified in Table 1, and the proposed sites are indicated on the map in Figure 1. Sponsors include utility companies and the associated research institute, utility vendors, state agencies, and an American Indian tribe. The proposed locations include six states: three along the Ohio River, one in the southeast, one on the Atlantic coast, and one in New England.

Table 2 provides a brief description of each project indicating the technology demonstrated, the size of the demonstration, the byproduct produced, and the estimated project cost. The selected projects include two full-scale demonstrations of limestone wet scrubbing to produce gypsum as the byproduct. Another is a biomass wet scrubbing technology that produces potassium sulfate and distilled water as byproducts. There are also two duct injection projects using calcium or sodium as reagents, one full-scale and the other at 5 MW, and one limestone furnace injection project that is about 33 MW in size. These processes all result in a dry solid waste for disposal. Finally, sulfur dioxide in a flue gas stream equivalent to 35 MW is catalytically oxidized to produce sulfuric acid in one project that was selected. The costs for these seven projects range from about \$9 million to \$37 million with the costs for the largest project not reported. Additional information about each of these projects is provided in the following section of the paper.

#### 100-MW DEMONSTRATION OF INNOVATIVE APPLICATIONS OF TECHNOLOGY FOR COST REDUCTIONS TO THE CHIYODA THOROUGHbred-121 FLUE GAS DESULFURIZATION PROCESS ON HIGH-SULFUR, COAL-FIRED BOILERS (SOUTHERN COMPANY SERVICES, INC.)

The Chiyoda Thoroughbred-121 (CT-121) Process is a second-generation limestone wet scrubbing process for removal of sulfur dioxide from utility boiler flue gas. The process incorporates forced oxidation to produce gypsum as the byproduct. The CT-121 process uses a unique absorber design called a Jet Bubbling Reactor (JBR). The JBR combines limestone dissolution, oxidation of calcium sulfite solids, and gypsum crystallization in one vessel. The flue gas is bubbled through the limestone slurry in the JBR, and as a result, the large recycle pumps used in conventional limestone wet FGD systems are not required. A very high quality gypsum that easily dewateres is produced. This gypsum can be disposed of using techniques developed by the phosphate industry and tested during a 20-MW prototype evaluation of the CT-121 process that was conducted at Gulf Power's Plant Scholz in the early 1980's. Based on this evaluation, commercial operation in Japan, and economic evaluations performed by the Southern Electric System, this technology was determined to be flexible, reliable, and cost competitive. However, cost reductions are reportedly possible through the application of innovative technology to the CT-121 process. The innovative applications include: 1) the use of fiberglass-reinforced plastic (FRP) and elimination of a prescrubber; 2) elimination of flue gas reheat; 3) elimination of a spare absorber; and 4) simultaneous particulate and SO<sub>2</sub> removal. The costs of a retrofit application are reported to be reduced by over 23% as a consequence of these innovative applications, and with a new installation, the reduction may be as much as 50% if the ESP or baghouse can be eliminated. Under the CCT II program, a 100-MW demonstration of these innovative applications has been proposed at Georgia Power Company's Plant Yates Unit 1 near Atlanta, Georgia.



DEMONSTRATION PROGRAM FOR POST-COMBUSTION DRY SORBENT INJECTION TECHNOLOGY  
(COMBUSTION ENGINEERING, INC.)

Three dry sorbent injection technologies are proposed for demonstration at Virginia Power Company's Yorktown Unit 2. The technologies to be demonstrated at the 180-MW unit include in-duct injection, in-duct spray drying, and convective pass injection (also called economizer injection). The basis for these technologies is the reaction of a calcium-based reagent with sulfur dioxide to produce calcium sulfite and calcium sulfate as a dry byproduct. The reagent is injected into the convective pass of the furnace or into the duct between the air preheater and the particulate collection device. The calcium sulfite and sulfate and fly ash are collected in an ESP or baghouse. The objectives of the proposed CCT II project are to demonstrate reductions in sulfur dioxide emissions at the Yorktown plant by 50% or more using these technologies and to provide data for the technical, economic, environmental, and operating evaluations of these technologies to support their commercialization.

COMMERCIAL DEMONSTRATION OF WSA-SNOX TECHNOLOGY AT OHIO EDISON'S NILES STATION  
(COMBUSTION ENGINEERING, INC. AND SNAMPROGETTI USA, INC.)

The WSA-SNOX Process was developed by Haldor Topsoe A/S, Denmark. It is a catalytic process for simultaneous removal of sulfur oxides and nitrogen oxides from utility boiler flue gas. Sulfur dioxide is oxidized catalytically to sulfur trioxide and recovered as concentrated sulfuric acid. The nitrogen oxide is reduced to free nitrogen by catalytic reduction with ammonia. This process can remove 95% of the sulfur oxides and about 90% of the nitrogen oxides in flue gas, producing a 93% to 97% concentrated sulfuric acid. The gas is first sent through a baghouse to remove particulates, then heated in an exchanger by the exiting gas stream to the temperature required for the catalytic reactions. Ammonia is added to the flue gas prior to the  $\text{NO}_x$  reactor where the  $\text{NO}_x$  is catalytically converted to nitrogen and water. Gas exiting the  $\text{NO}_x$  reactor is further heated, processed through a  $\text{SO}_2$  reactor, and catalytically converted to  $\text{SO}_3$ . This  $\text{SO}_3$ -laden gas is then cooled by heat exchange with the  $\text{NO}_x$  reactor feed gas and sent to a condensing tower where high concentration sulfuric acid is formed.

The basic technology development for the WSA-SNOX process began in Europe in the 1960s. During the past eight years, Haldor Topsoe A/S has developed the WSA-2 tower, an air-cooled falling film condenser of a unique design which is integrated into this process. The first commercial unit was installed in Sweden in 1986 to treat a 1.5%  $\text{SO}_2$  stream to achieve 95% removal and produce a 95% to 96% sulfuric acid byproduct.<sup>2</sup> The SCR catalyst was also developed by Haldor Topsoe A/S in work performed in 1983-1985. Testing has been conducted with a 6300-cfm demonstration unit under high and low dust loadings treating flue gas from a coal-fired power plant. The combination of the  $\text{NO}_x$  and  $\text{SO}_2$  removal steps is designated as the WSA-SNOX Process and has been undergoing testing since 1985. A 3-MW test unit which was started up in late 1987 in Denmark provides additional input for the unit proposed under the CCT II program.

The CCT II project will treat flue gas equivalent to about 35 MW of coal-fired power generation from Ohio Edison's Niles Station Unit 2. Unit 2 burns a coal with a typical sulfur content of 3.8% and produces a flue gas containing approximately 700 ppm  $\text{NO}_x$ . There are no reagents required for sulfur dioxide control and the byproduct produced is concentrated sulfuric acid. This technology has been applied to a pulp mill off-gas in Taiwan and a petroleum refinery coke-burning electric power plant in Italy. A U.S. coal-fired utility demonstration is needed prior to application of this technology to the utility industry in the U.S.



#### 5-MWe DEMONSTRATION OF THE $\text{SO}_x$ - $\text{NO}_x$ - $\text{RO}_x$ BOX (SNRB) PROCESS (BABCOCK & WILCOX)

The SNRB Process accomplishes the removal of  $\text{SO}_2$ ,  $\text{NO}_x$ , and particulates in one unit (a high-temperature baghouse). Dry injection of a calcium- or sodium-based reagent into the flue gas achieves the  $\text{SO}_2$  removal. The  $\text{NO}_x$  is reacted with ammonia in an SCR system upstream of the air preheater. Finally, particulate removal is accomplished in front of the air preheater using a high-temperature baghouse. The site-specific application determines the choice of the  $\text{SO}_2$  sorbent. High-sulfur applications in the East are likely to use calcium due to the lower cost of the reagent and of waste disposal relative to sodium. The more reactive sodium sorbents may be more attractive for Western sites where once-through operation may be more economical.

Babcock & Wilcox has proposed installation and operation of a 5-MWe demonstration project at Ohio Edison Company's R.E. Burger Unit 5 in Dilles Bottom, Ohio. The demonstration unit will be treating flue gas from a boiler firing Ohio bituminous coal. Operational and performance data will be obtained to evaluate the SNRB Process and to determine any impact of the technology on boiler and plant operation. The overall objective is to demonstrate the readiness of this technology for commercial application. Other specific objectives for the demonstration project are 70% to 90%  $\text{SO}_2$  removal at cost-effective sorbent-to-sulfur ratios, 90%  $\text{NO}_x$  removal at cost-effective ammonia-to- $\text{NO}_x$  ratios, 99+% particulate removal, integration of flue gas cleaning into a single-unit operation, and reuse or safe disposal of the byproducts.

The potential advantages for the SNRB Process that have been highlighted by the process developer include high  $\text{SO}_2$ ,  $\text{NO}_x$ , and particulate removal in one-unit operation; increased efficiency due to reduced flue gas temperatures and recovery of heats of reaction from pollutant removal; and production of a dry byproduct for disposal.

#### PASSAMAQUODDY INNOVATIVE CLEAN COAL TECHNOLOGY PROGRAM APPLICATION (PASSAMAQUODDY TRIBE)

The Passamaquoddy Tribe of Maine has developed a retrofit wet scrubbing technology for removing pollutants from the flue gas at coal-burning facilities. Specifically, the "Recovery Scrubber" reduces  $\text{SO}_2$  emissions by over 90%, uses a waste product as the reagent, and produces saleable by-products (potassium sulfate fertilizer and distilled water). The Recovery Scrubber was developed as the result of research and development efforts at the Tribe's Dragon Products Company, a cement manufacturer. While investigating methods for reclaiming potassium from the cement plant waste dust, it was determined that  $\text{SO}_2$  was scrubbed from the flue gas from the coal-fired cement kiln. The gas was contacted with a solution containing cement dust as the scrubbing reagent. Further research led to the discovery that biomass ash could be substituted as a reagent for applications of the Recovery Scrubber outside the cement industry. The reacted biomass ash which will be the only byproduct from the process requiring disposal is reported to be more benign as a result of its use in the scrubber.

The unit at the Dragon cement plant will be designed to demonstrate 90%  $\text{SO}_2$  removal while treating gas from a coal-fired cement kiln. The primary objectives of the project are to verify the  $\text{SO}_2$  removal efficiency and to obtain additional information on the economics associated with this technology.

## ADVANCED ON-SITE FLUE GAS DESULFURIZATION PROCESS (PURE AIR)

Air Products and Chemicals, Inc. and Mitsubishi Heavy Industries America, Inc. have formed a joint venture company, Pure Air, to market wet limestone FGD technology in the U.S. Pure Air has been selected under the CCT II program to install an advanced wet limestone FGD system at Northern Indiana Public Service Company's (NIPSCO's) Dean H. Mitchell Station. The FGD system will treat flue gas from multiple boilers with a total capacity of about 529 MW. The byproduct from the process will be gypsum that can be sent to disposal or used to make wallboard. The objectives of the project are to demonstrate innovative concepts that will reduce the capital cost of the FGD system and also result in operating costs that are substantially lower than those experienced currently.

The innovative features of the FGD system include: 1) use of a single 100%-capacity scrubber module to treat flue gas from multiple boilers with a capacity of 529 MW; 2) a single-scrubber loop with in-situ oxidation using a new air rotary sparger system; and 3) a new wastewater evaporation system (WES) to achieve zero water discharge. Demonstration of the WES will promote its use with new and existing FGD systems. NIPSCO will also be firing a variety of high-sulfur coals from Indiana, Illinois, or Kentucky to evaluate the flexibility of the FGD system to operate with a range of flue gas compositions.

## LOW NO<sub>x</sub>/SO<sub>2</sub> BURNER RETROFIT FOR UTILITY CYCLONE BOILERS (TRANSALTA RESOURCES INVESTMENT CORPORATION)

TransAlta Resources Investment Corporation is a wholly owned non-regulated subsidiary of the largest investor-owned utility in Canada, TransAlta Utilities Corporation. TransAlta's proposal to demonstrate and document the operational and economic performance of the Low NO<sub>x</sub>/SO<sub>2</sub> Burner (LNS Burner) on a utility cyclone boiler was selected under the CCT II program. A 33-MW cyclone boiler of Southern Illinois Power Cooperative near Marion, Illinois will be retrofitted with LNS Burners. The boiler will have two LNS Burners rated at 200 MBtu/hr each that will burn a total of about 10 tons of Illinois high-sulfur coal each hour. The LNS Burner is reported to provide SO<sub>2</sub> and NO<sub>x</sub> control within the burner through an innovative combustion process. Calcium reagent is injected through the burner to accomplish the SO<sub>2</sub> control. The burner which operates at a high temperature also causes much of the coal ash to be removed as molten slag.

The performance goals for the LNS Burner are 90% SO<sub>2</sub> control and an emission rate of 0.1 lb/MBtu (100 ppm) for NO<sub>x</sub>. This performance satisfies that required for bituminous coal-burning facilities by the New Source Performance Standards (NSPS) for SO<sub>2</sub> and surpasses the NSPS limit on NO<sub>x</sub> emissions of 0.6 lb/MBtu. TransAlta acquired this technology from Rockwell International in 1986 and has proceeded with commercialization. As part of these efforts, a single full-scale burner will be evaluated firing Western low-sulfur subbituminous coal. This demonstration will be located in Alberta, Canada and is expected to start in 1989.

## SO<sub>2</sub> AND COMBINED SO<sub>2</sub>/NO<sub>x</sub> CONTROL RESEARCH

The Department of Energy, Illinois, Ohio, and private organizations are also sponsoring a significant research effort in the areas of SO<sub>2</sub> control and combined SO<sub>2</sub>/NO<sub>x</sub> control. As mentioned previously, DOE in-duct sorbent injection technology development activities are continuing at a rapid pace. Extensive bench- and pilot-scale work continues in support of the development and evaluation of new process concepts and sorbents. Much of this work is summarized in the paper to be

presented by DOE in Session 8, Post-Combustion Dry Technologies, of this symposium. As a result, this research will not be covered in this paper.

A variety of other research activities that are sponsored by DOE, Illinois, Ohio, and private organizations is summarized in Tables 3 and 4 and is further discussed in the following paragraphs.

A 40-MW-equivalent CT-121 system has been installed at the University of Illinois to treat the gas from three stoker boilers which burn local high-sulfur coal. The CT-121 process is a wet limestone forced-oxidized system that produces high-quality gypsum as a byproduct. The objectives of the program are to closely monitor process operations for one year so that a detailed evaluation of the performance and cost of the CT-121 can be made and compared to other competing technologies.

The E-SOX Process is a concept developed by EPA for retrofit to utility boilers. An existing ESP is modified to incorporate lime slurry atomization in the front portion with the calcium sulfite/sulfate solids and fly ash collected in the remaining fields of the ESP. Modifications to upgrade the ESP will be required in most applications. A pilot test program is being cosponsored by Ohio, Babcock & Wilcox, and EPA. Session 8 of this symposium contains a paper providing information about the status of E-SOX research.

The Ohio Coal Development Office (OCDO) and several universities are sponsoring more developmental research at laboratory-scale and bench-scale. These projects are being conducted at the Universities of Cincinnati, Tennessee, and Toledo. In one project, lime spray drying is combined with ammonia injection to achieve 90%  $\text{SO}_2$  removal. The other three projects are focusing on novel approaches to removing  $\text{SO}_2$  from flue gas. One approach is to contact the flue gas with a dry sorbent in a fluidized bed reactor operated at high gas velocities. The use of ion-exchange resins in a fixed-bed reactor is another concept being examined. The flue gas must be cooled prior to  $\text{SO}_2$  absorption, and the loaded resin would be thermally regenerated. Finally, research is being conducted on a hollow-fiber membrane contactor that would be an alternative to a spray or packed tower for  $\text{SO}_2$  removal. The objective is to reduce absorber costs and offer more flexible operation.

Research activities related to combined  $\text{SO}_2/\text{NO}_x$  control are focused on developing additives to conventional wet and dry flue gas desulfurization systems and advanced flue gas cleaning processes which have high removal efficiencies for both  $\text{SO}_2$  and  $\text{NO}_x$  in one process. Research activities for the most part have been supported by DOE and industry.

Argonne National Laboratory (ANL) has an ongoing program to evaluate and develop additives for conventional wet and dry FGD systems. The ANL has completed testing of the addition of caustic soda (NaOH) to the lime slurry feed of their 20-MWe spray dryer. They report achieving over 50%  $\text{NO}_x$  reduction. ANL has also conducted bench-scale testing of different metal chelates. They report very high (90%) removals of  $\text{SO}_2$  and  $\text{NO}_x$  with Fe(II) EDTA and Al-citrate in a wet scrubbing system.

Pilot-scale testing of three high removal efficiency  $\text{SO}_2/\text{NO}_x$  control processes has been conducted in the U.S. The NOXSO process has achieved 90%  $\text{SO}_2/\text{NO}_x$  removal on the 0.75-MWe unit operated at DOE's Pittsburgh Energy Technology Center. Based on pilot-scale test results, bench-scale testing of the fluid bed copper oxide process is being conducted by Universal Oil Products (UOP) under funding from DOE. To date, 90%  $\text{SO}_2$  reduction and 77%  $\text{NO}_x$  reduction have been achieved.



Design of a 3-MWe pilot unit is underway. The DOE has also funded process development research on the E-Beam process. The results of the small pilot-scale testing at the Stout Power Plant in 1983-84 have been used to develop a 100-MWe design and an economic analysis of the process. Research and development efforts related to the Strategic Defense Initiative have resulted in significantly lowering the cost of electron beam generators.

The Ohio Coal Development Office and Columbia Gas System Service Corporation are working together to develop the Parsons Process for flue gas  $\text{SO}_x$  and  $\text{NO}_x$  control. Natural gas is used in this process to catalytically reduce  $\text{SO}_x$  to sulfur and  $\text{NO}_x$  to nitrogen. The project is currently in Phase II which includes procurement, erection, and testing of a pilot unit treating a flue gas slipstream from a coal-fired boiler. One of the primary objectives of the pilot testing is to demonstrate 90% reductions in both  $\text{SO}_x$  and  $\text{NO}_x$  emissions.

In summary, the majority of the research and development activities ongoing in the U.S. are focused on the following areas:

- Demonstration of clean coal technologies;
- Development of low-cost retrofit controls;
- Development of advanced  $\text{SO}_2$  and  $\text{SO}_2/\text{NO}_x$  control processes; and
- Improvement in the reliability and performance of controls applied to existing boilers.

The level of activity is extensive, as indicated by the variety of topics discussed at this symposium and the significant level of involvement of government agencies, research institutes, and private industry.

#### DISCLAIMER

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.



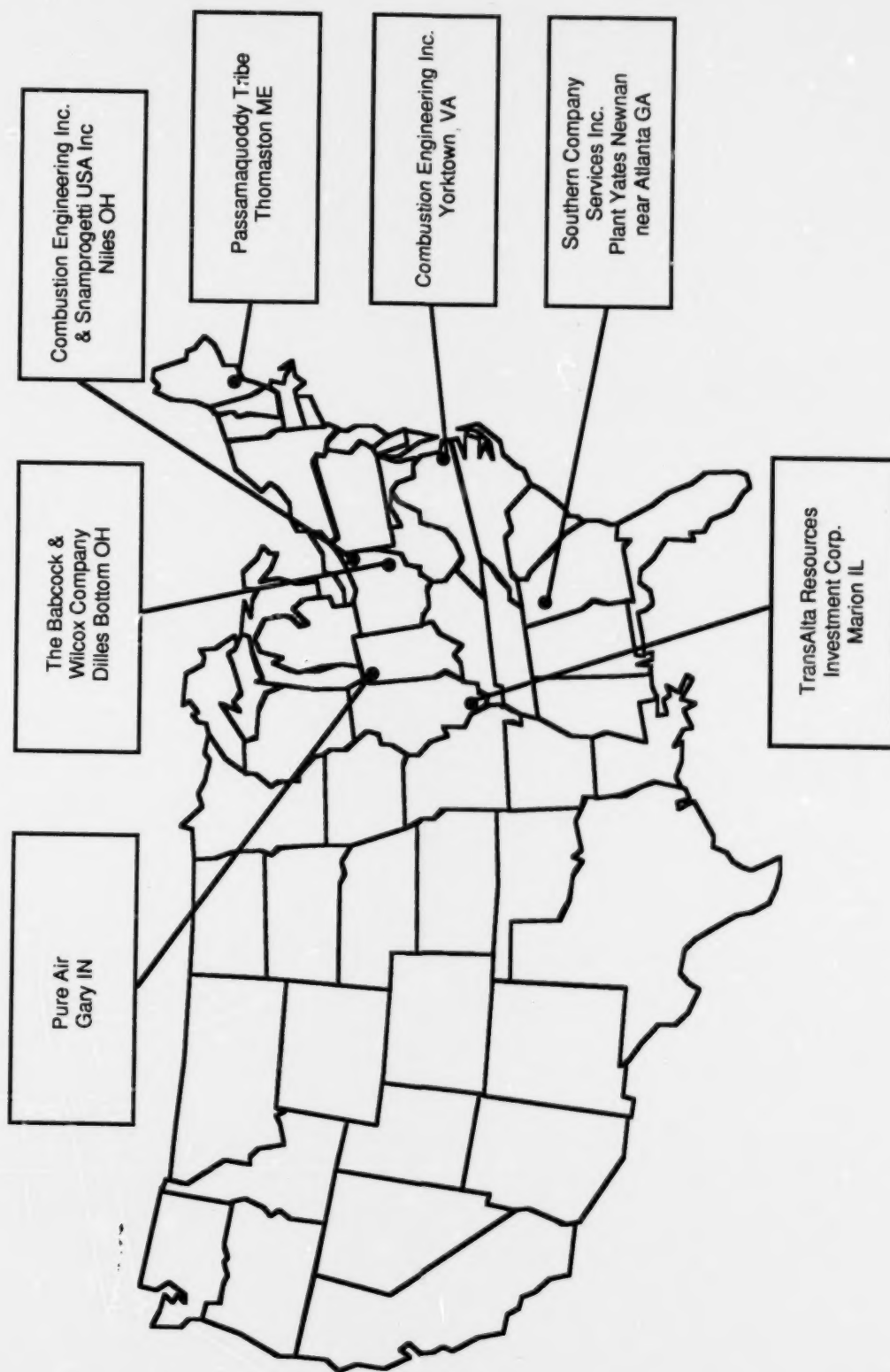


Figure 1. Clean Coal Technology SO<sub>2</sub> control demonstrations.

TABLE 1. CCT II WET AND DRY FGD PROJECTS

| Sponsors                                                                                                                                 | Project Title                                                                                                                                                                                          | Location                                                              |
|------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------|
| Southern Company<br>Services, Inc.<br>Electric Power<br>Research Institute                                                               | 100-MW Demonstration of Innovative<br>Applications of Technology for<br>Cost Reductions to the Chiyoda<br>Thoroughbred 121 Flue Gas Desul-<br>furization Process on High-Sulfur,<br>Coal-Fired Boilers | Plant Yates<br>Georgia Power Co.<br>near Atlanta, GA                  |
| Combustion Engi-<br>neering, Inc.<br>Virginia Power Co.<br>Electric Power<br>Research Institute                                          | Demonstration Program for Post-<br>Combustion Dry Sorbent Injection<br>Technology                                                                                                                      | Yorktown No. 2<br>Virginia Power Co.<br>Yorktown, VA                  |
| Combustion Engi-<br>neering, Inc.<br>Ohio Edison Co.<br>Snamprogetti<br>Ohio Coal Develop-<br>ment Office                                | Commercial Demonstration of WSA-<br>SNOX Technology at Ohio Edison's<br>Niles Station, Weathersfield<br>Township, Niles, Ohio                                                                          | Niles Station<br>Ohio Edison Co.<br>Niles, OH                         |
| The Babcock &<br>Wilcox Co.<br>Ohio Edison Co.<br>Ohio Coal Develop-<br>ment Office<br>Electric Power<br>Research Institute              | 5-MWe Demonstration of the SO <sub>x</sub> -<br>NO <sub>x</sub> -RO <sub>x</sub> Box (SNRB) Process                                                                                                    | R.E. Burger No. 5<br>Ohio Edison Co.<br>Dilles Bottom, OH             |
| Passamaquoddy Tribe                                                                                                                      | Passamaquoddy Innovative Clean<br>Coal Technology Program Applica-<br>tion                                                                                                                             | Dragon Products<br>Co.<br>Thomaston, ME                               |
| Pure Air<br>Northern Indiana<br>Public Services<br>Co.                                                                                   | Advanced On-Site Flue Gas Desul-<br>furization Process                                                                                                                                                 | D.H. Mitchell Sta.<br>Northern Indiana<br>Public Services<br>Gary, IN |
| Trans Alta Resources<br>Investment Corp.<br>Illinois Dept. of<br>Energy & Natural<br>Resources<br>Southern Illinois<br>Power Cooperative | Low NO <sub>x</sub> /SO <sub>x</sub> Burner Retrofit for<br>Utility Cyclone Boilers                                                                                                                    | Southern Illinois<br>Power Coop.<br>Marion, IL                        |

TABLE 2. CCT II FGD PROJECT DESCRIPTIONS

| Project Title                                                                                                                                                                         | SO <sub>2</sub> Control Technology                                                      | Size   | Byproduct                                                  | Project Cost (\$million) |
|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------|--------|------------------------------------------------------------|--------------------------|
| 100-MW Demonstration of Innovative Applications of Technology for Cost Reductions to the Chiyoda Thoroughbred 121 Flue Gas Desulfurization Process on High-Sulfur, Coal-Fired Boilers | Limestone wet scrubbing                                                                 | 100 MW | Gypsum                                                     | 35.8                     |
| Demonstration Program for Post-Combustion Dry Sorbent Injection Technology                                                                                                            | In-duct injection, in-duct spray drying, and economizer injection with calcium reagents | 180 MW | Dry solids (calcium sulfite, calcium sulfate, and fly ash) | 37                       |
| Commercial Demonstration of WSA-SNOX Technology at Ohio Edison's Niles Station, Weathersfield Township, Niles, Ohio                                                                   | Catalytic oxidation                                                                     | 35 MW  | Sulfuric acid                                              | 30                       |
| 5-MWe Demonstration of the SO <sub>x</sub> -NO <sub>x</sub> -RO <sub>x</sub> Box (SNRB) Process                                                                                       | Dry injection with calcium and sodium reagents                                          | 5 MW   | Dry solids (calcium or sodium sulfate and fly ash)         | 10                       |
| Passamaquoddy Innovative Clean Coal Technology Program Application                                                                                                                    | Biomass ash wet scrubbing                                                               | ---    | Potassium sulfate and distilled water                      | 9.4                      |
| Advanced On-Site Flue Gas Desulfurization Process                                                                                                                                     | Limestone wet scrubbing                                                                 | 529 MW | Gypsum                                                     | ---                      |
| Low NO <sub>x</sub> /SO <sub>x</sub> Burner Retrofit for Utility Cyclone Boilers                                                                                                      | Limestone furnace injection                                                             | 33 MW  | Dry solids (calcium sulfite, calcium sulfate, and fly ash) | 13.6                     |

TABLE 3. SO<sub>2</sub> CONTROL RESEARCH SUMMARY

| Application                                           | Development Status | Status and Summary of Results                                                                                                                  | Sponsors                                      |
|-------------------------------------------------------|--------------------|------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------|
| Chiyoda Thoroughbred-121 Process                      | 40-MWe             | Acceptance test completed; monitoring program underway; complete results in late 1989                                                          | Illinois, EPRI                                |
| E-SOX                                                 | 5-MWe              | 50% SO <sub>2</sub> removal achieved in lab tests; pilot tests are scheduled to begin in 1989                                                  | Ohio, EPA, Babcock & Wilcox, DOE              |
| Circulating Fluid Bed Absorber                        | Bench-scale        | Testing of the CFB reactor to examine low-temperature sodium bicarbonate; 90% SO <sub>2</sub> removal with 5-inch water pressure drop achieved | Ohio, Univ. of Cincinnati                     |
| Combined Lime/Ammonia Spray Dryer                     | Bench-scale        | Testing is ongoing; achieved 95% removal at 0.8 Ca/S ratio and 0.25 NH <sub>3</sub> /S ratio                                                   | Ohio, Univ. of Cincinnati, Univ. of Tennessee |
| Adsorption on Fixed Bed Resins                        | Lab-scale          | Single- and multi-component testing to determine resin adsorption capacity completed                                                           | Ohio, Univ. of Cincinnati                     |
| Hollow-Fiber Contactor for SO <sub>2</sub> Absorption | Bench-scale        | Bench testing is in progress with 90% SO <sub>2</sub> removal achieved using a sodium sulfite solution                                         | Ohio, Univ. of Toledo                         |



TABLE 4. COMBINED SO<sub>2</sub>/NO<sub>x</sub> CONTROL RESEARCH SUMMARY

| Application                                            | Development Status | Status and Summary of Results                                                                                                                 | Sponsors                                                 |
|--------------------------------------------------------|--------------------|-----------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------|
| NaOH Additive to Spray Dryer                           | 20-MWe pilot       | 50% NO <sub>x</sub> removal achieved with >70% SO <sub>2</sub> removal.                                                                       | DOE, ANL                                                 |
| Metal Chelate Addition to Wet Scrubber                 | Bench-scale        | Up to 90% NO removal using Fe(II) EDTA and Al-citrate                                                                                         | ANL                                                      |
| NOXSO Process                                          | 0.75-MWe pilot     | Achieved 90% SO <sub>2</sub> and NO <sub>x</sub> removal; 5-MWe pilot unit under construction                                                 | NOXSO, DOE                                               |
| Fluid Bed Copper-Oxide                                 | Bench-scale        | Achieved 90% SO <sub>2</sub> and 77% NO <sub>x</sub> removal; design of 3-MWe pilot unit underway                                             | UOP, DOE, Illinois                                       |
| Electron Beam                                          | Pilot testing      | 80-KW process demonstration unit operated in 1983-84 at Indiana Power and Light's Stout Plant; 100-MWe design and economic analysis completed | Ebara, DOE                                               |
| Parsons Process - Catalytic Reduction with Natural Gas | Pilot plant        | Engineering and cost studies completed; pilot evaluation in progress                                                                          | Ohio, Columbia Gas, R.M. Parsons, Delmarva Power & Light |

A PROGRAMME FOR REDUCING SO<sub>2</sub> EMISSIONS  
FROM U.K. POWER STATIONS - PRESENT AND FUTURE

W.S. Kyte,  
Corporate Environment Unit,  
Central Electricity Generating Board,  
Courtenay House,  
18 Warwick Lane,  
London, EC4P 4EB, U.K.

ABSTRACT

In this Paper the background to the programme, which the Central Electricity Generating Board is undertaking to reduce its sulphur dioxide emissions, is detailed. Factors which influence the types of technologies and processes to be used are discussed together with the disposal of the products from these processes. The role of advanced coal combustion plant is also discussed.

INTRODUCTION

There are four utility companies responsible for the generation of electricity in the United Kingdom which have a total installed capacity of just under 70,000 MW.

The Central Electricity Generating Board (CEGB) is responsible for the generation and transmission of bulk electricity in England and Wales and has a total of 74 power stations with a declared net capacity of 54,000 MW of which 67% is coal, 16% oil, 9% nuclear and the remainder gas turbine, hydro and pumped storage. Coal is the predominant fuel with over 80% of electricity being generated by the combustion of some 80 million tonnes of coal per annum.

The consumption of coal on this scale raises numerous important issues, some of which are environmental. The Central Electricity Generating Board has a statutory obligation to ensure that the effects of its operations on the environment are properly taken into account and it maintains a large research programme to ensure that any environmental impact of coal-fired generation is quantified and controlled within acceptable limits (1).

## CEGB VOLUNTARY ABATEMENT PROGRAMME

Acidification of the environment has been a growing concern over the last decade. In certain areas of Scandinavia and Scotland it has led to the loss of fish stocks from lakes. A major contribution to acidification of soils and waters is the emission of sulphur dioxide and nitrogen oxides from the combustion of fossil fuels.

The Central Electricity Generating Board has contributed significantly to the study of the acidification phenomenon, both by its own research and, in partnership with British Coal, by supporting financially a major study carried out by the Royal Society in collaboration with the Scandinavian Academies.

By early 1986, it had become clear that the process of acidification was of long standing; probably dating from the start of the Industrial Revolution. It was also probable that recovery of affected soils and waters would be slow, even if deposition of sulphur and nitrogen compounds was reduced. Nevertheless, the rate of deposition should not be allowed to increase and should be reduced as soon as practicable. The Board therefore reviewed its expected future emissions with the aim of taking the appropriate measures.

Emissions of sulphur dioxide from CEGB power stations have fallen in recent years and there will be continuing significant reductions in emissions from this source as the present generation of power stations is progressively replaced by plant incorporating "acid free" technology, i.e. nuclear power or fossil-fuelled stations equipped with high efficiency sulphur removal equipment. The first of this new generation of plant is expected to be commissioned in the mid-1990's, with emission reductions from this source becoming increasingly significant around the turn of the century, and beyond. To ensure continuation of the recent reduction in sulphur emissions (despite increasing levels of electricity demand) the U.K. Government, in response to a request by the Central Electricity Generating Board, has authorised the Board to retrofit some 6000 MW of coal-fired power plant - equivalent to three major power stations - with flue gas desulphurisation (FGD) plant. At the same time, the CEGB announced that it would be fitting FGD to any new coal-fired power stations.

Since 1985, the CEGB has been carrying out a series of trials of low NO<sub>x</sub> burners on various boiler types and in 1987 it announced a ten year programme to retrofit all its large bituminous coal-fired boilers with low NO<sub>x</sub> burners. This involves the replacement of 2000 burners in 44 boilers with a total generating capacity of 23,000 MW.

The FGD retrofit programme will start with the 4000 MW (6 x 660 MW) power station at Drax which when completed will be the largest FGD plant in the world. Consent has just been granted for this development after long planning negotiations and the first site contract has just been let. It is expected that the main FGD contract will be let by the end of 1988 and the plant will commission in phases - two units in 1993, two in 1994 and the remaining two in 1995.

## U.K. AND EEC REGULATIONS

In January 1988, H.M. Inspectorate of Pollution, who are responsible for licencing new power plant, issued a new edition of their Notes on Best Practicable Means (BPM) for boilers larger than 700 MW<sup>th</sup> which codify the regulations (2). These call for a 90% removal of sulphur dioxide based on a 30 day rolling average for all new plant. The Notes also specify a nitrogen oxide limit of 650 mg/m<sup>3</sup> (6% O<sub>2</sub>). These limits are only applicable to new plant, older plant being allowed to continue to operate under the previous regulations where limits were only specified for particulate emissions.

In June 1988 Environment Ministers of the EEC, meeting in Luxembourg, reached agreement on a Directive on The Limitation of Emission of Pollutants Into The Air From Large Combustion Plants commonly known as the Large Combustion Plant Directive. This Directive sets limits for both new and existing plants for all plants greater than 50 MW<sup>th</sup>. The regulations for new plant are complex but consist essentially of a variable sulphur dioxide emission limit of 2000 mg/m<sup>3</sup> (6% O<sub>2</sub>) at 100 MW<sup>th</sup> falling to 400 mg/m<sup>3</sup> at 500 MW<sup>th</sup> for coal-fired boilers. For high or considerably fluctuating sulphur content in indigenous coals, a percentage removal is specified rising from 40% at 100 MW<sup>th</sup> to 90% at 500 MW<sup>th</sup>. For nitrogen oxides the limit, for coal-firing, is 650 mg/m<sup>3</sup> (6% O<sub>2</sub>). Limits are also set for oil and gas firing.

In a U.K. context, it can be seen that for new plants the BPM and LCP requirements are identical.

The LCP Directive also calls for reductions from existing plants. However, this reduction is not plant based but is applied across the whole class of plant. Each country in the EEC has agreed under this Directive to reduce its sulphur dioxide and nitrogen oxide emissions from this class of plant by various percentages of the 1980 emissions by specified dates. For the U.K., the stipulations are that it should reduce its sulphur dioxide emissions from existing plant in three phases - 20% by 1993, 40% by 1998 and 60% by 2003. For nitrogen oxides, the reductions are 15% by 1993 and 30% by 1998.

The implications of this Directive for the CEGB are not yet clear but it is probable that the equivalent of an extra 6000 MW of existing plant, in addition to the voluntary programme, will need to be fitted with flue gas desulphurisation. This, together with the expected new plant building programme, will mean that about 20,000 MW of FGD plant will be operating or under construction by the year 2000.

It is expected that the voluntary low NO<sub>x</sub> retrofit programme will enable the NO<sub>x</sub> requirements of the Directive to be met.

The FGD retrofit programme must be seen in conjunction with major construction programmes for new CEGB nuclear and coal-fired plant which will take place over the same period of time. Construction has just started on a new 1300 MW PWR nuclear plant and an application for consent has been lodged for another PWR and sites are being investigated



for future PWR consent applications. The CEGB has also applied for consent to construct two new coal-fired power stations and preparations are in hand to apply for consent for an additional coal-fired power station. It is proposed that these stations would be constructed to the new CEGB reference design with two 900 MW units for each plant. All the stations would have burners designed to reduce NO<sub>x</sub> emissions and would be fitted with FGD plant to the appropriate standards.

#### CHOICE OF FGD SYSTEMS AND STATIONS

As part of a long programme of work into the most cost effective methods of reducing sulphur dioxide emissions, the CEGB has examined many options ranging from conventional coal cleaning, advanced coal cleaning, through limestone injection into the boiler to flue gas desulphurisation. The results of these studies have been published elsewhere (3) and hence only a summary will be given here. The studies showed that there was only very limited scope for coal cleaning since most of CEGB coals are already cleaned for ash removal. A detailed study (4) of limestone injection showed that the system would be very expensive compared with wet FGD systems on the CEGB's large 2000 MW base-load coal-fired stations. For these reasons, the CEGB initiated detailed studies of FGD systems.

There are several factors, which are unique to the CEGB system, which have had a marked influence on the way in which FGD will develop in the U.K. These are:-

- (i) The bulk of generation is centred on large coal-fired power stations.
- (ii) All units on the same site are of similar age.
- (iii) The high chloride content of U.K. coals.
- (iv) Very tight timescale allowing no scope for pilot or demonstration programme. The CEGB may have up to 16,000 MW of FGD under construction before the first unit is commissioned.

In addition to these factors, several criteria determine the type of FGD plant which is suitable. These criteria are:-

- (i) It must be well proven and commercially available on a large scale.
- (ii) Adverse side effects must be reduced to the minimum.
- (iii) The FGD product should preferably have a socially beneficial use.

When these factors and criteria are taken into account, then the only suitable processes are the limestone/gypsum, regenerative, spray-dry and seawater systems. Of these, only the two former are

suitable for the present programme of FGD for the early retrofits and new stations. The seawater process may, after development, be suitable for a later new coastal or estuarine site whilst spray-dry is most suited to low load factor or smaller plant.

For new stations, site specific factors relate to the choice of the FGD system though this could have an influence on the original choice of the site. For retrofit purposes, the choice of FGD system and station are inter-related and may require to be chosen together. Site specific factors may also be influenced by overall global factors such as the marketing and disposal of FGD products.

The choice of stations for retrofit is governed by many parameters among which the most significant are:-

- (i) Sulphur levels in coal and coal burn.
- (ii) Life of station and load factor.
- (iii) Ease of fitting of FGD.
- (iv) Local product marketability and availability of disposal sites.
- (v) Environmental factors.
- (vi) Supply of raw materials.
- (vii) Costs and economics.

The first FGD retrofit will be installed at Drax Power Station in Yorkshire which will be fitted with a limestone/gypsum process producing commercial quality gypsum for sale into the wallboard industry. Similarly, the first two new stations at W. Burton 'B' and Fawley 'B' will use the limestone/gypsum process and produce commercial quality gypsum for sale. The sites and processes for the other retrofits have not yet been decided.

#### DISPOSAL OF PRODUCTS

The CEGB has a declared policy of putting the products of combustion to socially beneficial use. It has carried out this policy in respect of ash for many years and in 1986/7 some 43% of the total ash production was sold of which about half went into the manufacture of lightweight insulating bricks. The majority of the unsold ash was used for land reclamation or beneficial landfill.

The combined new and retrofit FGD programmes have logistical implications for raw materials supply and by-product disposal which are of considerable importance and complexity. Each 2000 MW power station equipped with limestone/gypsum FGD technology will require about 1/3 million tonnes per annum of limestone and will produce around 1/2 million tonnes per annum of gypsum. The latter represents about 15 - 20% of the

current U.K. market for this product. Similarly, a 2000 MW station equipped with regenerative FGD technology would produce some 4 million tonnes per annum of sulphuric acid. Though this is only about 10-12% of the U.K. market for sulphuric acid, there is only very limited scope for the installation of regenerative FGD plant since the products must be integrated into a local chemical market. Marketing of the products from the total FGD programme cannot be assured. Environmentally acceptable routes for disposal of by-products must be identified and secured well in advance of FGD operations. Similar consideration has to be given to the quarrying and transportation of limestone for use in FGD plants. Local communities must be assured that the effects on their environment during construction and operation of these plants will be acceptable.

It is the intention to market as much of the products of FGD as possible in order to minimise the quantities requiring disposal. The CEEB has an extensive research programme to ensure that it produces gypsum of the right quality to be suitable for use in the construction industry.

The CEEB is also undertaking a major research programme to establish other options for the disposal of surplus gypsum. These include landfill, land reclamation, artificial reefs and use as a land restorative for saline soils.

#### ADVANCED COAL COMBUSTION PLANT

In recent years, considerable research and development effort has been devoted to assessing various alternative "advanced" coal combustion technologies based on fluidised bed combustion or coal gasification. These have been considered to be potential competitors to pulverised fuel combustion with flue gas desulphurisation for power generation around the turn of the century. They lend themselves to more efficient operation in combined cycle mode, which incorporates a gas and a steam turbine. The desulphurising plant required is very much smaller and hence cheaper, and the basic units may be amenable to factory fabrication. However, there are other factors that may offset the potential cost savings. For example, fluidised beds have a higher limestone requirement than FGD units on conventional plant for the same sulphur removal.

The CEEB and British Coal have recently completed a £28M joint research programme at the Grimethorpe Pressurised Fluidised Bed Research Establishment. The programme has investigated many aspects of the technology, in particular ways of controlling erosion of boiler tubes in the fluidised bed - a process once thought to threaten the economics of this type of plant.

Completion of this joint programme coincides with the completion of two design studies of alternative advanced coal conversion systems for large scale electricity generation in the U.K., and the results have just been published by the U.K. Department of Energy as Energy Paper No. 56 (5). In one of the studies, the CEEB collaborated with British Coal

and the Department of Energy on an engineering assessment of combined cycles based on fluidised bed combustors and in the second with British Coal, British Gas and the Department of Energy on the application of the British Gas/Lurgi slagging gasifier to large scale combined cycles when the necessary gas turbines become commercially available in the 1990's.

The study suggests that at the large plant sizes traditionally of interest to the CEGB, the coal gasification route has the greater potential for generating electricity more cheaply and with lower emissions to the atmosphere than conventional plant fitted with emission controls.

Smaller units using any of the technologies were shown to produce electricity at higher cost unless overall efficiency can be significantly increased, as for example in a CHP application. At smaller unit sizes, fluidised bed combustion might be a better option than conventional plant fitted with FGD.

#### ACKNOWLEDGMENT

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#### DISCLAIMER

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.



**Progress in European SO<sub>2</sub>-NO<sub>x</sub>-Control**  
**An Overview**

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by Dr. Bernhard Forck  
VCB Technical Association  
of Large Power Plant Operators  
Essen, Federal Republic  
of Germany

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Mailing address of the author:  
V C B  
Postfach 10 39 32  
D-4300 Essen 1  
Federal Republic of Germany

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Progress in European SO<sub>2</sub>-NO<sub>x</sub>-Control  
An Overview

Nations are complicated constructions.

The more complicated is Europe with more than 30 different nations having mostly their own languages,

their own histories,  
their own traditions,  
their own legal systems,  
their own media, and public opinions  
their own economic and social systems,  
and different geography and wealth.

The European nations are organized in several associations such as UN-ECE, OECD, Comecon, EEC. Comecon may represent the "eastern" part, EEC the "western" part of Europe; another part is Scandinavia, of which Denmark is member of the ECE, and of which Finland cooperates with (but is not member of) Comecon. Further nations are Austria, Switzerland, Yugoslavia and others.

If one concentrates on the EEC:

There are twelve nations,

the smallest:

Luxembourg, 2,600 km<sup>2</sup>, 0.4 million inhabitants,

the largest:

France, 544,000 km<sup>2</sup>, 52.2 million inhabitants,

all together

2,254,000 km<sup>2</sup>, 313.3 million inhabitants

(USA 9,372,600 km<sup>2</sup>, 239.3 million inhabitants),

with nine different, more or less related languages (Portuguese - Spanish - Italian - French, Greek, Danish - German - Dutch - Englisch, the latter also with relations to French). These nations are on the way to a union and will suspend their common borders in 1992: the real Common Market. There is some common

legislation, guidelines, which have to be or are transformed into national legislation, a long lasting process.

As examples may be stated guidelines for SO<sub>2</sub>, particulate matter and NO<sub>x</sub> ambient concentration limits, and limits for trace elements in drinking water, or, latest and with respect to SO<sub>2</sub> and NO<sub>x</sub> control with power stations, the guideline for large firings. This guideline constitutes emission limit values for new "energy producing" combustion plants licensed after July 1, 1987, for particulates, SO<sub>2</sub> and NO<sub>x</sub> as follows in Table 1. These emission value limits are - roughly - defined as 48-hour-means if continuously measured. There are no emission value limits set for existing plants, that means those in operation or licensed before July 1, 1987. For existing plants there are emission ceilings agreed upon in three phases for SO<sub>2</sub> (until 1993, 1998, 2003, respectively) nation by nation and in two phases for NO<sub>x</sub> (until 1993, 1998, respectively). The total emission reduction in the EEC will be from

|                                       | 1980 | 1993                     | 1998   | 2003   |
|---------------------------------------|------|--------------------------|--------|--------|
| 14.4 million t/a                      |      | SO <sub>2</sub> : 23.3 % | 41.8 % | 57.4 % |
| 3.7 million t/a (as NO <sub>2</sub> ) |      | NO <sub>x</sub> : 10.7 % | 29.8 % | *)     |

The model for this EEC guideline was the German "Großfeuerungsanlagenverordnung", abbreviated as GFAVO or referred to as 13. BImSchV, which in 1983 set stronger emission value limits for new and existing plants. The total contents of this complex regulation cannot be described here, but one essential example should be given (in mg/m<sup>3</sup> of flue gas) <sup>\*\*) :</sup>

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)\* t/a means metric tons per annum

)\*\* m<sup>3</sup> means normal cubic meter at  
standard conditions, 1,013 mbar, 273 K, dry

|                                       | dust | SO <sub>2</sub> | NO <sub>x</sub>             |
|---------------------------------------|------|-----------------|-----------------------------|
| new coal fired power station          |      |                 |                             |
| thermal capacity 300 MW <sub>th</sub> | 50   | max. 400        |                             |
| dry bottom firing:                    |      |                 | 800 (6 % O <sub>2</sub> )   |
| slag tap firing:                      |      |                 | 1,800 (5 % O <sub>2</sub> ) |

The prevailing SO<sub>2</sub> emission limitation is 15 % of the fuel sulphur.

These emission limitations have been valid also for existing plants (which are not scheduled for shutdown until April 1, 1993) as of July 1, 1988. Less than one year after the release of the GFAVO the eleven state ministers responsible for environmental protection in the Federal Republic agreed to distinctly lower NO<sub>x</sub> limits, in the given example 200 mg/m<sup>3</sup>, to comply with as soon as possible, that means end of 1989 or of 1990.

Let me come back to this regulation later in this paper.

There are similar emission value limits set in The Netherlands and in Austria, the latter a non-EEC-member. Some other EEC-members have higher SO<sub>2</sub> limits or limitate the fuel sulphur content only, some have no limits for coal firings like France or Great Britain. NO<sub>x</sub> limits are still rarer. More active in the field of SO<sub>2</sub> and NO<sub>x</sub> limitation are the Scandinavian countries.

All these "active" countries as a consequence of their geographical and climatic situation accept more pollutants by long-range transport than they deliver to their neighbour countries! The Comecon-members, centrally planned economies, or Yugoslavia feel being too poor in capital for air pollution control installations with power stations.



Another word regarding Great Britain and France. Though Great Britain has set no general emission limit values, the English CEEB is planning retrofitted FGD plants for 6,000 MW power station capacity to come on stream after 1993 and to equip 2,000 MW new capacity with FGD. Low  $\text{NO}_x$  burners should be installed if possible.

France on the other hand intends to rely more or less fully on nuclear energy; fossil fueled power plants will be shut down, so that there will be fossil fueled boilers only in the industry boilers. The final result is a  $\text{SO}_2$  and  $\text{NO}_x$  emission reduction "by other means". In other EEC countries like Denmark or Italy there are strong political forces against nuclear energy!

All this may give an idea about the in-homogeneity in Europe at all and even in the EEC, and about the difficulty to give a short survey of EUROPE, and may it be only in the field of air pollution control.

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The technical basis for  $\text{SO}_2$  and  $\text{NO}_x$  control with power stations has been laid very early. We know the lime operated scrubbers in London since 1938. Early tests with active coal or coke  $\text{SO}_2$  absorption or with limestone injection were made in Germany in the 50's. No doubt the first "commercial" FGD plants have been installed in the USA by several suppliers, f.i. Combustion Engineering, Babcock and Wilcox, or the former Chemico. In 1974 VGB organized a trip to US and Japanese FGD plants for the judgement, whether FGD might be "state-of-the-art". At that time we learned about the operation difficulties especially with corrosion, scaling or plugging in plants

where dust and  $\text{SO}_2$  removal were combined, and about the large lagoons for the sludge disposal. We saw the magnesium oxide and the Catox process, in Japan the Chiyoda process, processes which disappeared from the market. Let me - very late - say many thanks for the insights we got at that time.

Another VGB trip to the USA at the end of the 70's resulted in the knowledge of the first attempts with low NO<sub>x</sub> combustion, f.i. BOOS or OFA.

At the early 80's we recognized the development of the catalytic  $\text{NO}_x$  reduction in Japan, which may have its basis on a German patent for  $\text{NO}_x$  reduction in the off-gas of a nitric acid plant.

So, by exchange of experience and by licences the SO<sub>2</sub> and NO<sub>x</sub> control technology has been introduced into Europe. Nevertheless, the first commercial FGD plants at power stations were proprietary processes of European suppliers. But others followed using US and Japanese know-how.

**Spray drying is a European development.**

.....

It is not easy to describe the progress or to find out, where and what progress is, in front of an assembly of experts who deal with a technology for a very long time.

As explained earlier, in the FRG the "Large Furnace Ordinance" (GFAVO, 13.BImSchV) has been issued in 1983 (and in Austria a respective law (DKEG)). To comply with it, any new power plant (there were only few ones) and any existing one with a thermal

capacity above 300 MW must be fitted with an effective ( 85 %) FGD as of July 1, 1988, at the latest, and with a DENOX unit until 1989 or 1990 (depends on agreements with the state authorities) in order not to exceed 200 mg/m<sup>3</sup>. Similar conditions are valid in Austria. Only power stations which are scheduled for shutdown until 1993 are excepted. Heavy fuel oil combustions may be converted to light heating oil in order to avoid the installation of FGD and sometimes also of catalytic DENOX.

#### SO<sub>2</sub> removal

As the 1st of July is gone in the FRG there are now in operation or in the last phases of construction 135 scrubbers or scrubbing units with a capacity of about 40,000 MW<sub>e</sub> (power stations for public service and several power plants in industries), of which about 85 % (by capacity) produce gypsum, 8 % spray dried powder. The remainder produce sulfur, sulfuric acid, ammonium sulfate, and a mixture of ashes after addition of calcium hydroxide into the combustion chamber (dry additiv process). Most of these units are retrofit installations and had to be planned, constructed and commissioned within the short period of five years. There was not enough time for principally new developments.

The German power stations' SO<sub>2</sub> emissions will go down from 1.55 million t/a in 1982 to 0.53 million t/a in 1989 and to 0.34 million t/a in 1993, that means to 35 and 22 %, respectively.

As mentioned, most of the applied processes are gypsum producing ones. One reason is that there is only few space for disposal areas; the intention by law is to avoid wastes as far as possible. The other reason is that wet scrubbing

with lime or limestone is the most efficient and economic process available and is experienced as reliable. It is now accommodated also to the conditions of the Rhinish lignite (or brown coal) which is burnt with its about 50 % water content. That means a higher part of the flue gas as normal is water vapour, and consequently, the scrubber temperature is much higher, 65 to 70 instead of 40 - 50 °C, which means less SO<sub>2</sub> solubility but higher reaction velocity.

Only few older plants use lime, most of them take limestone as the absorbens. Because of the buffer capacity limestone eases the pH control which is critical. Normally the pH of the scrubber inlet liquid is kept at pH 6 to 5.5; the pH falls to about 3.5 to 4 at the scrubber sump enhancing the sulfite to sulfate oxidation. This oxidation is completed by air blown into the scrubber sump (forced oxidation), some separate oxidation vessels are bypassed now. For this air introduction several devices have been developed with different results. One of the best measures is the air distribution by or the vicinity of agitators if there are ones.

Well operated forced oxidation lime or limestone scrubbers now have an SO<sub>2</sub> removal efficiency of 90 % +. The reliability or availability of the FGD plants equals - after the usual debottlenecking - the reliability or availability of the boilers. Several parts are stored as spare parts, f.i. pump wheels, valves, control and monitoring devices, but there is no spare scrubber installed even not with the base load lignite power stations. By GFAVO it is quoted for a scrubbed flue gas temperature of 72 °C or more at the exit of the stack in order to avoid condensates and droplet rain near the power station. Several reheat devices are used. In most cases Ljungström-type heat exchangers are installed. Different constructions are used like special sealings



and a purge gas section, so that unscrubbed flue gas does not enter the clean gas, in order not to deteriorate the overall scrubber efficiency. To suppress plugging of the heat exchanger by droplet evaporation the clean gas is reheated by several degrees C by recycling a slip stream of the clean gas. A new arrangement is to install the necessary fan on the cold, wet side of the scrubber. The compression heat adds to the flue gas preheating in front of the heat exchanger. Only few plugging and corrosion on the fan blades has been experienced.

Other reheat systems are:

- heat transportation, that means two separate tube and shell heat exchangers with circulation of water between them,
- mixing hot air from an additional flue gas - air preheater (also Ljungström type) into the clean gas
- mixing recycled steam reheated clean gas into the cold clean gas
- reheat by high pressure steam only.

The latter arrangement was chosen in a special case due to space problems.

In one experimental power plant the total FGD unit is placed inside a cooling tower; the cleaned gas is carried away together with the cooling tower plume. By monitoring ground level  $\text{SO}_2$  concentrations and by observing the plume's behavior in the lower atmosphere from an air plane it could be proved that the pollutants' dispersion is better than from a considerably higher chimney. This arrangement now is used in many of the retrofitted lignite power stations and is also planned for others. There is no need for sophisticated heat exchangers but for separate emergency stacks, used during

the firings' start-up, and if the FGD is bypassed (which is lawful for 72 hours in series and 240 hours per year). The cooling towers' inner walls have to be especially protected against corrosion.

Corrosion and erosion are sometimes still unsolved problems. For pumps new alloys have been developed, there is a lot of (not only good) experience with rubber and plastic linings of tubes and scrubbers. Also brick work is used as a lining, especially at the flue gas entrance. In some units tubes with smaller diameter in four special cases also the scrubbers are made of fibre reinforced polyesters.

As the emission values are legally defined as half hour averages (which might be exceeded in few cases) and daily averages (which must not be exceeded), the industry now manufactures reliable monitoring instruments and apparatus for the statistical evaluation of the measurements.

With gypsum producing FGD plants there is a water blow-down in order to keep the liquors' chloride concentrations constant. This waste water originally is supersaturated with gypsum and contains trace elements from the remainder of particulates downstream the ESP (mostly below 50 mg/m<sup>3</sup>) and to a greater extend from the lime or limestone (remember: the circulating liquors have a pH below 6, in pre-scrubbers below 1). According to the law the waste water must not be released into rivers or lakes uncleaned, there have to be make-up units. By treating these water flows with lime or caustic soda gypsum and trace element hydroxides are precipitated, with the aid of flocculants the waste water is clarified. There are concentration limits set, f.i. for COD 150 mg/l, Cd and Hg 0.05 mg/l (eventually reached by additon of sulfides), Pb 0.1 mg/l, Zn 1 mg/l.

The sulfate concentration is kept below 2,000 mg/l. The residual filter cake is disposed of or used as a cement additive.

The FGD gypsum is separated from the mother liquor by means of centrifuges or, in more modern plants, by vacuum drum or belt filters. The moisture content is normally below 10 or 7 %. This fine powder is stored in silos or halls and shipped as such. In some plants the gypsum powder is dried and mechanically compacted to briquettes, about 3 cm in diameter. These briquettes can be stored outside, they are easier to handle in the gypsum processing industry, similar to naturally mined gypsum. About two thirds of the German FGD gypsum will be processed industrially this ways. The remainder, most from the lignite firings, will be mixed with the strong alkaline ashes and the blow-down water, and then disposed of in the lignite strip mines. The particular disposal areas are fitted with groundwater protections.

Some of the German and Austrian coal-fired power stations use the spray drying FGD process with good success. There is a number of suppliers offering several process variations as far as the spraying is concerned, f.i. co- or countercurrent gas - spray flow, lime suspension distribution by rotating cups or by pressurized air, bag filters or ESP's used for reclaiming the sulfur compounds containing powder. Part of this powder is recycled to the lime suspension with the consequence that a desulphurization efficiency of more than 85 % with a stoichiometric Ca/S ratio of 1.2 to 1.5 is achieved. There is no need for reheat or or waste water make up.

The great draw back of this easier to handle and more economic process is the low availability of disposal area. The authorities are afraid, that sulfates, chlorides and trace elements could be

leached out by rain (though the stuff is alkaline and can solidify with the air-borne  $\text{CO}_2$ ). So up to now this FGD residue can only be disposed of underground (f.i. in coal or salt deep mines) or in thoroughly tightened pits.

There is research under way to apply this residue in construction materials (bricks, concrete) or elsewhere, but we are still waiting for practicable results.

Some other FGD plants apply the Wellman-Lord process ( 8 scrubbers) for sulfur and sulfuric acid production, having their own problems with the by-product sodium sulfate; one ammonia scrubbing yielding ammonium sulfate, one catalytic SO<sub>2</sub> conversion to sulfuric acid, and one active coke adsorption are installed. Both the latter are combined with a DENOX stage.

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Many successful attempts have been made to reduce the  $\text{NO}_x$  formation during combustion. Multistage burners have been developed; the over fire (and under fire) air technology, fuel staging, in-furnace-reduction, and flue gas recycling are used. Even with slag tap firings and with gas-turbines combustion modifications resulted in partly remarkable lowering of the flue gas  $\text{NO}_x$  concentrations. Generally the - higher - emission value limits of the GFAVO are met, in cases of oil and gas firings also the lower ones. Only few oil fired boilers, nearly none with gas firing therefore are to be retrofitted with a separate DENOX unit.

Most of the firings fueled with Rhinish lignite (brown coal) meet the lower NO<sub>x</sub> limits by means of fuel and air staging, and



in-furnace-reduction. Only few of them will be equipped with additional installations, presumably ammonia or urea injection.

All combustion modifications had to be adjusted to the details of the individual firings or boilers.

With the ministers' decision to set the emission value limit at 200 mg/m<sup>3</sup> for coal firings (150 mg/m<sup>3</sup> for oil and 100 mg/m<sup>3</sup> for gas firings) to be reached "as soon as possible" a hectic activity must be taken. In a very short time about 60 small test units were planned and erected, yielding valuable results. Most of these test facilities contained different available Japanese catalysts, which have been tested at oil and gas firings, only few at coal firings, these operating in base load. Catalysts and processes had to be adapted to German (=Austrian) operating conditions, that are: medium and peak load, other sorts of coal, combination with existing FGD, dry bottom or slag tap firing, and others. Very early two large power stations, at that time being under construction, could be reconstructed by insertion of the necessary catalyst boxes.

The first catalysts for the test and "commercial" units were bought in Japan, but soon after there began German catalyst manufacturing after Japanese licences or after own developments. The positive effect is that the prices per ton of catalyst went down.

One of the most essential findings were the effect of arsenic lowering the catalysts' activity. Arsenic occurs in higher concentration in the flue gas of slag tap firings if the ESP fly ash is - more or less - fully recycled into the combustion chamber. This brought up the idea to arrange the catalytic DENOX

(referred to as SCR) downstream of the FGD, though there is the need of flue gas reheating from about 50 to between 320 and 400°C by gas-gas heat exchange and additional heat input (gas fire or high pressure steam). Now there are offered arsenic-resistant catalysts.

The - few - SCR units now in operation comply with the emission value limits. But there is a lot of work to be done for the first start-up. A particular essential is the adjustment of an equal ammonia to  $\text{NO}_x$  distribution in order to have the designed stoichiometric ratio of nearly unity everywhere across the duct. Measurement of the ammonia slip is only possible by time wasting wet chemical methods.

With dry bottom firings, of which the fly ash is delivered to the concrete consumers as an additive, the ammonia slip after the catalysts is to be kept lower by a factor of ten than the 5 ppm the Japanese suppliers recommend. Only in this way the ammonia content of the fly ash can be kept below the 50 ppm the construction industry asks for.

In these cases there is not great a danger to plug the air preheater with ammonium bisulfate or to contaminate the FGD liquors. Nevertheless installations are on hand for air preheater washing.

It is too early to give a general report on operation experience because most of the DENOX units upstream of the ESP or downstream of the FGD - the latter not only at slag-tap firing, but also dictated by space problems in retrofitting cases - are still under construction.

It should be mentioned, that the DENOX catalysts not only accelerate the  $\text{NO}_x$  reduction, or better, the ammonia oxidation but also, to a certain degree, the  $\text{SO}_2$  oxidation. By this the acid dew point of the flue gas increases giving rise to higher corrosion rates.

By now the installation of DENOX units for a capacity of 32.000 MW is planned, by far SCR processes of different licensors. It is foreseen, that the  $\text{NO}_x$  emissions out of the German power stations for public service will be lowered from

0.75 million t/a in 1982 to  
0.58 million t/a in 1987 and to  
0.20 million t/a at the beginning of the 90's,

that is finally to less than 30 %. This includes combustion modifications.

In some power stations tests were made with the ammonia injection into the flue gas at temperatures between 1,000 and 900 °C. The results were encouraging for further application at one or another boiler. Analogous results yielded tests with urea solution; a first installation is ordered.

One unit just started up took the active coke process for simultaneous removal of  $\text{SO}_2$  and  $\text{NO}_x$ . There have been some problems with the internal coke transportation, but the  $\text{NO}_x$  removal is satisfying.

In the context with the said DENOX test units several attempts are made to develop other processes for simultaneous  $\text{SO}_2$  and  $\text{NO}_x$  removal, f.i. the electron beam process which, had promising

results, the EDTA process which could not be applied commercially because of efficiency and waste water problems, the ozone process, which suffered from high ozone generation costs and from low solution velocity of the  $\text{NO}_2$ , or a process, which imitates the a century old lead chamber sulfuric acid process in combination with NO oxidation by air, yielding nitric acid. All these processes were not far enough developed at the time when decisions for application must be made. So now there are only few incentives for further work with these processes though they have some advantages from an environmental particularly by-product point of view.

#### Acknowledgement

A great deal of information and facts laid down here are made available orally in VGB conferences and committee meetings, others are taken from widely spread references most of them being in German language. So I decided not to add a long reference list. If there is interest in special facts or single references, please contact the author.



Table 1  
EEC guideline: Emission value limits, mg/m<sup>3</sup> (m<sup>3</sup>: 1,013 mbar, 0°C, dry, reference O<sub>2</sub> concentration)

| Fuel:                | solid                                               | liquid                      | gaseous                      | remarks                                      |
|----------------------|-----------------------------------------------------|-----------------------------|------------------------------|----------------------------------------------|
| SO <sub>2</sub>      |                                                     |                             |                              |                                              |
| thermal heat input   |                                                     |                             |                              | thermal capacity between the limits: sliding |
| 100 MW <sub>th</sub> | 2,000 (6 % O <sub>2</sub> )                         | 1,700 (3 % O <sub>2</sub> ) | all 35 (3 % O <sub>2</sub> ) | the limits: sliding                          |
| 500 MW <sub>th</sub> | 400                                                 | 400                         | (LPG: 5 )                    | going over between                           |
|                      |                                                     |                             | 2000 and 400 (solid fuel)    |                                              |
|                      |                                                     |                             | 1700 and 400 (liquid fuel)   |                                              |
| NO <sub>x</sub>      |                                                     |                             |                              |                                              |
|                      | 650                                                 | 450                         | 350                          |                                              |
|                      | solid fuels with less than 10 % volatile compounds: |                             |                              |                                              |
|                      | 1,300                                               |                             |                              |                                              |
| Dust                 |                                                     |                             |                              |                                              |
| thermal heat input   |                                                     |                             |                              | blast furnace gas 10                         |
| 500 MW <sub>th</sub> | 100                                                 | all: 50                     | all: 5                       | gases from                                   |
| 500 MW <sub>th</sub> | 50                                                  |                             |                              | steel industry 50                            |

PROGRESS IN EUROPEAN SO<sub>2</sub>  
AND NO<sub>x</sub> EMISSION CONTROL

by: Helmut Weiler  
STEAG Aktiengesellschaft  
4300 Essen 1, FRG

and

William Ellison  
Ellison Consultants  
Monrovia, MD 21770

ABSTRACT

An updating is provided of SO<sub>2</sub> and NO<sub>x</sub> removal technology developments and utilization in the conduct of the substantial acid rain control program in Europe that began in West Germany in 1983/1984.

INTRODUCTION

Acceleration of European activities for reduction of SO<sub>2</sub> and NO<sub>x</sub> emissions since 1983 is centered in the Federal Republic of Germany, (West Germany), where retrofit SO<sub>2</sub> and NO<sub>x</sub> emission controls are now in place at electric utility plants and large boiler facilities. The strenuous program also calls for controls on industrial boilers, large and small in the early 1990s. This activity is the result of extensive forest damage, considered to be tied to air pollution, which has occurred in some regions of that and other countries. Effects on trees have been particularly severe in the highly industrialized area of North Rhine-Westphalia in the central portion of West Germany as well as in Baden Wuerttemberg in the southwest. Major SO<sub>2</sub> and NO<sub>x</sub> emission reductions are being carried out in a number of other countries throughout Europe in a parallel and continuing effort to control the environmental impact of acid precipitation. The West German electric utility projects deadlined for achievement of required SO<sub>2</sub> removal by July 1, 1988, have been finished on schedule. The completion during the period 1983-1988 of FGD retrofitting to an aggregate of 37,000 MW(e) of West German power plants, each of total capacity greater than approximately 110 MW(e), i.e. 300 MW(t), has reduced their annual SO<sub>2</sub> emission from 1.53 down to 0.53 million metric tons.

## COAL USE IN WEST GERMANY

### LOW SULFUR

Typical bituminous coal fired in West German boilers is comparatively low, i.e. 0.9 to 1.2% sulfur coal, and for this reason, in part, problems and cost of flue gas desulfurization operations are substantially less than those encountered in common high sulfur service in U.S.A. Nationwide primary energy consumption during the late 1980s equals approximately 11,000 PJ/annum, the full-load equivalent of 350 GW(t), approximately 30% (105 GW(t)) by coal. Nearly 2/3 of the coal use is by power plants but approximately 25% (26 GW(t) i.e. 26,000 MW(t)) is supplied for industrial use. Industry and many electric power plants generally use physically cleaned, high grade bituminous (hard) coal with lower heating value of 25,000 to 30,000 kJ/kg (10,800 to 12,900 Btu/lb.), medium volatiles content, 5-10% ash and 0.9 to 1.2% sulfur. Basis coal heating value of 11,850 Btu/lb, annual coal use by all sectors in West Germany equals 120 million metric tons and use by industry totals 30 million tons. However, 30% of coal fired in plants larger than 105 MW(e) size is lignite with sulfur content of 0.3-1.2%.

### HIGH SULFUR

Ballast coal (middlings from coal cleaning) with substantially higher sulfur emission is fired in a number of units by two large power companies, STEAG and VKR, each of which is a subsidiary of government-owned coal companies. For example STEAG fires approximately 2,000 MW(e) of capacity with ballast coal containing up to 1.4% sulfur, and up to 4,600 mg/Nm<sup>3</sup>, i.e. 1,650 ppmv, SO<sub>2</sub> in the raw flue gas. In some instances ballast coal fuel contains as much as 2.7% sulfur.

In some isolated instances lignite firing yields a very high flue gas SO<sub>2</sub> concentration.

## GOVERNMENT REGULATIONS

West German regulations issued in 1983 and 1984 address existing and new boilers of size 18 MW(e) and larger, impose stringent SO<sub>2</sub> and NO<sub>x</sub> controls on all boilers and plants above approximately 110 MW(e) i.e. 1,030 million Btu/hour heat input, and include requirements for retrofitting of FGD on units as small as 35 MW(e), equivalent to approximately 340 million Btu/hour. SO<sub>2</sub> and NO<sub>x</sub> control compliance above 18 MW(e), i.e. 170 million Btu/hour, is conservatively tied to half-hour mean values of emissions (1). This typically requires gas cleaning systems above 35 MW(e) size to be provided with design emission levels substantially lower than the regulatory emission-limit values. The 1986 Clean Air Act (TA Luft) (2), also referred to as TI Air, is a general administrative regulation for comparatively small industrial sources including coal fired boilers from 1/3 MW(e) to 18 MW(e) size, i.e. 3.4 to 170 million Btu/hour and, when invoked by local/regional jurisdictions on a case-by-case basis, imposes stringent SO<sub>2</sub> and NO<sub>x</sub> emission limits.

### SO<sub>2</sub> EMISSION CONTROL

In 1980, SO<sub>2</sub> emissions in West Germany totalled approximately 3.2 million metric tons, 1.9 million tons, i.e. 60%, of which was from power plants. The Emission Control Act (GFAVO) of 1983 (3), (Ordinance on Large Combustion Plants) has required that coal- or oil-fired units and individual plants larger than approximately 110 MW(e), aggregating over 40,000 MW(e) capacity, achieve 85% SO<sub>2</sub>

removal, and an SO<sub>2</sub> emission level of 400 mg/Nm<sup>3</sup> or 144 ppmv (approximately 0.3 lb/million Btu), whichever is more stringent, by July 1, 1988. However, based on other provisions of this national directive, boiler owners have designated specific units comprising 12,000 MW(e) for retirement by 1993 after no more than 30,000 additional hours of operation at an SO<sub>2</sub> emission rate not to exceed 2,500 mg/Nm<sup>3</sup> (2.0 lb/10<sup>6</sup> Btu). Units and plants in the range of 35 to 110 MW(e) are specified for 60% removal and 2,000 mg/Nm<sup>3</sup> or 720 ppmv (1.6 lb/10<sup>6</sup> Btu) by April 1, 1993. Units from 18 to 35 MW(e) are also required to meet 2000 mg/Nm<sup>3</sup> by April 1, 1993. There are approximately 400 coal-fired boilers in West Germany in the size range 35 to 110 MW(e). The TI Air lays down the principles of the licensing procedures for the smaller, industrial sources including coal-fired boilers of size 1/3 to 18 MW(e), and contains air quality standards for about 12 pollutants, general emission standards for all pollutants, calculation procedures for stack heights and pollutant distribution and provisions for monitoring and measurements. The SO<sub>2</sub> emission limit for coal fired boilers, new and existing, is 2000 mg/Nm<sup>3</sup>, (720 ppmv), 7% O<sub>2</sub> basis. Moreover, SO<sub>2</sub> reductions, such as by dry additive injection, will ultimately be required so as to decrease SO<sub>2</sub> emission by at least 50%. Fluid bed boilers must limit SO<sub>2</sub> emissions to 400 mg/Nm<sup>3</sup>, (144 ppmv), anticipating 75% SO<sub>2</sub> removal. These emission standards must be met within a time period of 3 to 8 years (from 1986), the specific deadline for individual sources depending on significance of the emission and the complexity of retrofit work. The formal manner of applying provisions of TI Air continues to be shaped as early implementation proceeds. There are approximately 3,900 coal-fired boilers in West Germany in the size range, 1/3 to 18 MW(e), 3600 of them smaller than 2 MW(e), i.e. 20 million Btu/hour, size.

#### NO<sub>x</sub> EMISSION CONTROL

In 1980, NO<sub>x</sub> emissions in West Germany totalled approximately 3.1 million metric tons, of which coal-fueled power plants accounted for 0.65 million tons. A 1984 accord among federal and provincial environmental ministers in West Germany requires that existing coal-fired units and plants larger than 110 MW(e) reduce NO<sub>x</sub> emissions to 200 mg NO<sub>2</sub>/Nm<sup>3</sup>, approximately 100 ppmv (0.163 lb./million Btu.) Units in the range of 18 to 110 MW(e) are specified for 650 mg/Nm<sup>3</sup> (325 ppmv), equivalent to 0.53 lb/million Btu, for dry bottom types, and 1,300 mg/Nm<sup>3</sup> (650 ppmv) for wet bottom (slag tap) types. (It is understood that these values have been subsequently reduced to 400 and 650 mg/Nm<sup>3</sup>.) Emission limits are on the basis of 6% O<sub>2</sub>, dry bottom, and 5% O<sub>2</sub> wet bottom. All existing boilers on a retirement schedule are allowed 650 mg/Nm<sup>3</sup> for dry bottom and 1,300 mg/Nm<sup>3</sup> for wet bottom. Compliance deadlines, set by individual state governments, generally specify no later than 1989 for dry bottom boilers and 1990 for wet bottom boilers. Limits are substantially lower (more stringent) for oil- and gas-fired boilers. TI Air imposes an NO<sub>x</sub> emission limit of 500 mg/Nm<sup>3</sup> (250 ppmv), equivalent to 0.42 lb/million Btu on coal fired boilers of size 1/3 to 18 MW(e) to be achieved by extensive combustion modification work. Fluid bed boilers must limit NO<sub>x</sub> emissions to 300 mg/Nm<sup>3</sup> (150 ppmv).

#### COMPARISON WITH NSPS (USA)

##### EPA-PROMULGATED NSPS FOR INDUSTRIAL BOILERS

Great attention has been given in the last year by industry in U.S.A. to requirements and means of SO<sub>2</sub> control for new coal and oil fired boilers in the size range, 29 to 73 MW(t), (100 to 250 million Btu/hour), specified by New Source Performance Standards. The standards promulgated by EPA in December, 1987 (4) call for 90% removal and an emission no greater than 1.2 lb per million Btu (0.8 for oil firing), substantially stricter criteria than presently apply to similar size



units, new or existing, in West Germany. (NSPS for boilers with less than 100 million Btu/hour heat input that could be expected to limit SO<sub>2</sub> emissions from coal firing to 1.2 lb./MM Btu are scheduled for promulgation by E.P.A. in 1990). See Table I displaying all EPA-promulgated NSPS values: SO<sub>2</sub>, NO<sub>x</sub> and particulate matter. However, the corresponding West German emission regulations, which additionally apply to existing units, are expected by private West German spokesmen to be adjusted to a more stringent level for both new and existing boilers, and the apparent difference in policy, U.S. vs. West Germany, would thereafter be correspondingly less. Moreover, the U.S. regulations apply to a 30-day rolling average emission and the West German values are tied to very short term (1/2 hour mean) emission measurements, (97% of which over a year's period must be smaller than 1.2 times the emission limit value) and thus, the difference in stringency between U.S. and West German requirements is substantially less than the numerical emission values would indicate. West German NO<sub>x</sub> emission standards for existing coal-fired boilers above 1/3 MW(e) size are more stringent than for new U.S. industrial boilers in the size range 10 to 26 MW(e). A comparison, West Germany vs U.S.A., for the full range of utility and industrial boiler sizes is given in Table II.

#### OTHER LEGISLATION IN EUROPE

Due to West German influence, Austria, Switzerland, Italy, The Netherlands, Denmark, Sweden, Finland, and the European Economic Community (EEC) have either enacted or proposed significant SO<sub>2</sub> and NO<sub>x</sub> control legislation. Major retrofit SO<sub>2</sub> control activity and emergence of NO<sub>x</sub> removal programs have been taking place, directed to utility size boilers, particularly in Austria, The Netherlands, Denmark and Sweden. Table III displays the NO<sub>x</sub> emission management objectives of governing officials for the EEC (5), including 1985 revisions, which anticipate emission limits for existing plants throughout Europe, after 1995, roughly comparable in severity to current nationwide requirements in West Germany. Objectives have since been restructured to establish 1993 and 1998 as deadline dates. With a unanimous vote of all member countries required for its passage, as has been the case, its approval has not been possible. However, new general EEC rules for legislation are pending which would require only a 2/3 majority of the member countries. Emission limits for large existing SO<sub>2</sub> sources are organized with 1993, 1998 and 2003 deadline dates. With EEC 1980 annual emissions of 14.4 million tons SO<sub>2</sub> and 3.7 million tons NO<sub>x</sub>, these programs would reduce NO<sub>x</sub> by 10.7% (1993) and 29.8% (1998), and SO<sub>2</sub> by 23.3% (1993), 41.8% (1998) and 57.4% (2003).

#### IMPACT OF FGD WASTE MANAGEMENT CONCERNS

##### AVOIDANCE OF SOLID AND LIQUID FGD WASTE

Unlike typical FGD practice to date for both utility and industrial boilers in U.S.A., process selection in Europe, especially in Germany, is substantially influenced by restrictive SO<sub>2</sub>-waste management considerations. Particularly with the utility boilers and largest of industrial boilers, production of a usable FGD byproduct is emphasized in West Germany due to severe constraints on throwaway solid-waste practices. Additionally, whereas use of throwaway sodium-base scrubbing is very common in industrial-boiler FGD systems in U.S.A., this approach is not workable in West Germany because of the substantial liquid effluent discharge of SO<sub>2</sub> waste. One exception is in the glass industry where recycling/recovery of the spent sodium alkali is expected to be possible.

## CONTRAST WITH U.S. INDUSTRIAL FGD PRACTICE

For purposes of comparison it is noted that of the more than 40 operating industrial boiler FGD systems in U.S.A. almost all are sodium-liquor scrubbing type, and in a few instances with lime post-precipitation to achieve dual alkali operation (6). The sodium-base scrubbers, while a highly efficient  $\text{SO}_2$  emission control means, generate and discard the  $\text{SO}_2$  catch as a saline liquid effluent rather than a precipitated solid mass, and require substantial receiving-stream assimilative capacity for dissolved solids disposal. In a comparable industrial application Europeans would design to yield a solid usable or throwaway waste.

## EMPHASIS ON GENERATION OF USABLE GYPSUM

In Europe, particularly in West Germany, management of  $\text{SO}_2$  wastes has been a major concern influencing choice of  $\text{SO}_2$  removal means, and will continue to be so in the impending program directed to large industrial boilers. Future broad application on large industrial boilers of commercial FGD technologies other than those that yield gypsum by-product will at the same time depend in great measure on the perceived ability to establish sustained use or acceptable means of disposal of quantities of future production of FGD solid waste. As a result of extensive retrofitting of lime/limestone scrubbing systems on utility boilers during the period 1984 to 1988, FGD gypsum production in West Germany including throwaway-grade gypsum from lignite fired units is expected to reach 2.5 million annual metric tons by 1988, with a substantially higher level by 1993 when FGD systems in the 35-105 MW(e) range are in operation. Extensive testing by the government of The Netherlands (where substantial FGD gypsum production is developing) of existing commercial sources of FGD gypsum in Germany has indicated that while this by-product differs in quality from one power plant source to another, broad commercial use (as gypsum) of diverse European FGD gypsum sources will generally be technically possible (7). An expanding European market for new gypsum sources can be expected since new process innovations may soon become available to economically convert significant amounts of FGD gypsum by-product to attractive anhydrite (anhydrous calcium sulfate) and the unique and valuable alpha hemihydrate (of calcium sulfate) forms. As per Table IV (8) ultimate commercial use of over five million annual metric tons of FGD gypsum has been forecast in West Germany.

The several hundred existing West German coal-fired boilers in the size range, 35 to 110 MW(e), that will require  $\text{SO}_2$  removal in the amount of 60% (or possibly substantially higher) by 1993 represent a very large ongoing FGD expenditure almost rivaling the cost of large utilities installations to meet the 1988 startup deadline. This former group consists primarily of bituminous-fired wet bottom boilers plus many bituminous-fired dry bottom boilers and lignite-fired boilers. If indications of an adequate gypsum market are believed and accepted, and particularly if control requirements are modified to call for  $\text{SO}_2$  removal efficiency substantially higher than 60%, thus further encouraging use of wet FGD, significant use of gypsum-producing limestone scrubbing systems of simplest single loop design would be expected to result. Such single-loop, in-situ forced oxidation FGD design has in the mid 1980s evolved in West Germany as the generic, industry standard for coal-fired electric utilities and is the most common single type of system purchased in Europe to date. Designed for and serving boilers with modest coal-sulfur concentrations, performance to date indicates good operating experience except for a minor pattern of mechanical problems with gypsum solids dewatering and handling facilities. Available information indicates that these systems reliably remove  $\text{SO}_2$  as required and yield gypsum by-product of quality suitable for use, with or without minor reprocessing, in manufacture of plasters, gypsum wallboard and cement. Most of the system installations of this type that will be in operation in Europe by 1990 are listed in Table V. However, if attractive FGD technologies such

as dry scrubbing including the common spray dryer type are found as expected to have a sufficient market for utilization of their solid waste by-product, or acceptable disposal means, many applications of these could occur. Furthermore, the normally preferred use of wet lime/limestone FGD so as to yield usable gypsum by-product generally calls for limiting of gypsum surface-moisture and chloride content, requiring essentially all of the chloride catch to be discarded as a liquid-effluent outfall. Inherently zero-effluent FGD means such as dry scrubbers therefore offer significant advantages in those instances where the dry scrubbing waste product can be satisfactorily managed. Most of the dry scrubbing installations in Europe that will be in operation by 1990 are listed in Table VI. Furthermore, conversion to natural gas or distillate oil fuel may be a feasible alternative to use of FGD in some cases, particularly in the 18 to 35 MW(e) size range.

## COMPARISON WITH THE U.S.A. AND JAPAN

### FGD SERVICE

As in Japan, most of the West German wet FGD installations are in low/medium sulfur coal service. Similarly, they are designed to employ forced oxidation to yield commercial by-product gypsum and thus avoid the substantial cost and complexity of throwaway management of solid waste. At the same time, application of wet FGD in West Germany differs significantly from that in the U.S. in some major respects:

- Limestone supply is generally ground at the mine source rather than at the boiler site.
- Due to comparatively high energy cost in Europe, scrubbed gas is reheated by scavenging heat from the FGD inlet gas. Rotary and tubular regenerative heat exchanger systems are being commonly employed in West Germany (avoiding need for augmented energy supply) to meet a mandatory 72°C (162°F) minimum stack outlet temperature requirement.
- Due to conservative FGD-buyer policy and the unusual stringency of stack emission control regulations tied to monitoring of half-hour mean values of emission rate, systems are frequently specified by the owner for a substantially lower stack emission rate than the GFAVO emission-limit value, e.g. SO<sub>2</sub> removal efficiency in excess of 90% is commonly provided for low-sulfur coal service.
- With typically low fuel sulfur level (e.g., flue-gas SO<sub>2</sub> concentration generally less than 3,000 mg/Nm<sup>3</sup>) and SO<sub>2</sub> removal efficiencies greater than 90%, extremely low acid-gas concentrations exist in the flue gas contacting the vulnerable, low-pH components downstream of the scrubbing sprays. Accordingly it has been possible to establish high (95%) availability system design without use of spare wet-scrubber modules.

### TYPES OF FGD SYSTEMS USED

Table VII illustrates basic differences and similarities in FGD practice in West Germany, Japan and U.S.A.



## OTHER WEST GERMAN SO<sub>2</sub> CONTROL INSTALLATIONS

### DRY SCRUBBING WITH SPRAY DRYER, THROWAWAY

A number of small and large European boilers aggregating over 5,000 MW(e), (a substantial number in West Germany) are being equipped with conventional throwaway-waste dry scrubbing systems using spray dryers employing lime, (Table VI). In instances where suitable solid waste management schemes are made available, this technique may be expected to be applied to additional existing West German boilers of size 110 MW(e) and less before the 1993 startup compliance deadline. European high sulfur coal applications of both the spray dryer and circulating-fluid-bed scrubber extend the electric utility use of these semidry FGD processes to coal sulfur levels substantially above those of present U.S. installations. The circulating fluid bed scrubber has also had significant European operating experience in this service, and a substantial basis for broader use of semi-dry scrubbing in the U.S. on high sulfur is developing. In FGD-using European countries other than West Germany, including Denmark, Austria, and Italy, conventional dry scrubbers are being applied at substantially higher inlet flue-gas concentrations (as high as 7000 mg SO<sub>2</sub>/Nm<sup>3</sup>) than earlier worldwide applications. In West Germany, commercial circulating-fluid-bed dry scrubbing is being applied, with semi-dry operation similar to that of spray dryers, at inlet flue-gas SO<sub>2</sub> concentrations as high as 13,000 mg SO<sub>2</sub>/Nm<sup>3</sup>. In some locations, particularly Sweden, spray drying systems are being installed downstream of high-efficiency fly ash collection equipment to more readily permit the FGD solid waste to be converted to a generally usable by-product.

### DRY SCRUBBING YIELDING ANHYDRITE

Using a system design originating in Sweden, two spray dryer type installations of approximately 100 MW(e) size have been installed downstream of electrostatic precipitators in West Germany with the aim of converting the FGD waste to anhydrite (anhydrous calcium sulfate) of a quality suitable for commercial use. However, a significant amount of HCl and HF are also collected and usability of the anhydrite is believed to be uncertain.

### CIRCULATING-FLUID-BED SCRUBBER

This hydrated-lime-using dry-scrubbing type process, the gas/liquid contactor of which is typically designed to operate upstream of a new or previously existing electrostatic precipitator, is unique and of potentially great significance for U.S., high-sulfur, throwaway-waste FGD applications because of the unusually high coal sulfur levels at which it is being applied (9). A field demonstration with a 50 MW(e) unit, that began in 1984 removed 95% SO<sub>2</sub> from a flue gas from lignite firing containing 4,250 mg SO<sub>2</sub>/Nm<sup>3</sup> (1500 ppmv), and a 125 MW(e) installation that started up in the past 12 months consistently achieves over 96% SO<sub>2</sub> removal in a low-grade lignite service with inlet flue gas containing 13,000 mg SO<sub>2</sub>/Nm<sup>3</sup> (4700 ppmv). The operating system achieves 95% availability and lime/sulfur stoichiometric ratio of 1.7 with the outlet gas temperature at a 27°F approach to the wet bulb temperature. In an upward flow dry/semi-dry reactor, inlet flue gas fed via an extended venturi nozzle is mixed with a finely pulverized hydrated lime and injected water. A relatively high flue-gas velocity (with system design values that can range between 6.5 and 26 ft/sec) carries unreacted lime and reaction products out of the top of the reactor. Later, solids are separated from the flue gas in the downstream particulate collector, typically a combination mechanical



collector and electrostatic precipitator, and a portion of it is recirculated to the reactor to significantly increase solids residence time and utilization.

#### AMMONIA SCRUBBING

A 240 MW(e) wet FGD installation in West Germany has operated for a number of years using ammonia scrubbing to produce ammonium sulfate by-product liquor that is converted to a low-grade, dry, fertilizer product in a small auxiliary spray-dryer system. Although highly publicized problems with a visible ammonia stack plume have for a time blocked completion of additional sales and installations, an engineering solution has been identified calling for use of high gas-pressure-drop mist eliminator equipment, and an additional installation of 75 MW(e) size is going forward. The ammonia scrubbing method for  $\text{SO}_2$  removal may become generally attractive because of its yield of a usable by-product other than gypsum.

#### LIME/LIMESTONE INJECTION INTO A MULTISTAGE BURNER

LIMB is of particular interest in Europe for retrofit emission control of industrial boilers. Principal worldwide application for LIMB to date is in lignite-fired electric-utility boilers in West Germany, where approximately 3000 MW(e) of existing lignite fired capacity was retrofitted by 1987 for temporary  $\text{SO}_2$ -emission-reduction duty in North Rhine-Westphalia (10). The optimum flue gas temperature range for  $\text{SO}_2$  removal is 800 to 1,100°C. Significant precipitator fouling problems and poor  $\text{SO}_2$  removal efficiency are reported for a full-scale 700 MW(e) test in Germany in bituminous coal fired service using limestone fed near the burners (11) but, generally, electrostatic precipitators have served adequately in sorbent injection installations in limiting stack emissions of particulates. In full scale tests at 125 MW(e) at another bituminous coal boiler in Germany limestone and hydrated lime reagent were fed alternately near the boiler-furnace outlet. With 2 moles of sorbent feed per mole of inlet  $\text{SO}_2$ , removal efficiency was 32% with limestone and 44% with hydrated lime. (Somewhat poorer results were achieved in limestone injection tests in a pulverized bituminous coal fired boiler in Malmo, Sweden). In short-term furnace injection tests in a small bituminous coal fired unit in Germany similar low removal efficiencies were measured (12). As a rule of thumb, a stoichiometry of 3 is needed for 60% removal. LIMB system design instead utilizing humidification of flue gas followed by dry lime injection is now considered to be more attractive, e.g. for large industrial boilers, since removal efficiency is significantly improved.

#### $\text{SO}_2$ CONTROL INSTALLATIONS IN OTHER EUROPEAN COUNTRIES

Influenced by this new control activity in Germany, approximately 2000 MW(e) of usable-gypsum-producing FGD systems are being installed in the Netherlands, and a major FGD installation program is underway in Austria on utility boilers. Other European countries including Sweden, Denmark, Italy, and England, have also become major markets for FGD, with many installations operating, under construction or planned. Many developments and installations are of possible significance to U.S. industrial boiler operators.

#### NORWAY

FGD has been installed on industrial sources in Norway such as aluminum smelters, industrial oil and coal fired boilers, and ilmenite pre-reduction furnaces and tail gas from Claus sulfur plants. A large FGD system is currently being supplied to a RCC-unit at an oil refinery. Nearly all the FGD systems are Flakt-Hydro seawater scrubbers aggregating the equivalent of 1430 MW(e) scrubber

capacity and including 4 industrial boiler applications in the range of 10 to 30 MW(e) put in service prior to 1983.

#### AUSTRIA

Many conventional wet scrubbing systems are installed. However, the first commercial system worldwide for boiler limestone injection went into operation at Voitsberg power plant, Unit 2, 65 MW(e), in lignite-fired service in 1983 (13). This retrofit installation achieves 57%  $\text{SO}_2$  removal. Three additional limestone injection systems aggregating 440 MW(e) have been installed. This includes a pilot plant installation on a boiler in bituminous coal service derated to 80 MW(e) for  $\text{SO}_2$  control by limestone injection at 80%  $\text{SO}_2$  removal. An 80%-efficient limestone injection system is installed on a 110 MW(e) lignite-fired boiler. A 133 MW(e) circulating fluid bed boiler has been installed at Gratkorn. This unit, the largest in Europe fires lignite and combustible wastes from pulp and paper production. Emission limits are 400 mg  $\text{SO}_2/\text{Nm}^3$  (140 ppm) and 300 mg  $\text{NO}_x$  as  $\text{NO}_2/\text{Nm}^3$  (150 ppm) but local authorities intend to lower the limit for  $\text{SO}_2$  to 200 mg/ $\text{Nm}^3$  (70 ppm) by this year, requiring increased grinding and rate of feed of limestone reagent.

#### SWEDEN

By the end of 1986 an aggregate of 1280 MW(t), i.e. approximately 470 MW(e), of dry-scrubbing FGD were in operation in low-sulfur coal service at the six district heating plants, some as small as 45-55 MW(e). Design removal efficiencies are generally in the range of 70 to 80% to achieve a 0.1 g S/MJ (570 mg  $\text{SO}_2/\text{m}^3$ ) emission limit value (14). Most of these installations include an electrostatic precipitator precollector, which will help ensure a maximum potential for utilization of the FGD solid waste. (Additionally, major progress has been made in gaining use of dry scrubber waste containing 10 to 30% fly ash). FGD operations at two of the plants began in 1982 and 1983 and has been quite good. In Sweden it has been shown that sulphur dioxide emissions of stoker fired boilers can be reduced by the feeding of limestone or dolomite above the burning fuel. This has been demonstrated at a plant comprising two 45 MW(e) boilers where it has been possible to achieve an  $\text{SO}_2$  removal efficiency of 60-70% with an emission limit of 0.1 g S/MJ (570 mg  $\text{SO}_2/\text{m}^3$ ), at a capital cost 35 to 50% of that of dry scrubbing installations of comparable capacity. The removal efficiency depends on the Ca/S molar ratio, the temperature in the furnace, and the retention time.

#### FINLAND

In Finland a 250 MW(e) limestone injection system (15) gains only 30% removal in the furnace but incorporates a tail end activation reactor (simplified spray type contactor) to humidify the flue gas upstream of the electrostatic precipitator to gain an attractive overall  $\text{SO}_2$  efficiency ranging from approximately 60% to over 80% with flue-gas temperature approach to the wet bulb temperature ranging from 36°F to 9°F, (20 to 5°C), respectively. The unit is in peaking service firing low-sulfur bituminous coal.

#### CZECHOSLOVAKIA

A wet magnesia scrubbing system of 200 MW(e) capacity yielding sulfuric acid by-product will begin operation on high sulfur lignite in 1989 (16). A number of power plants are being equipped with calcium-base systems, including limestone injection and wet scrubbing.

## FURTHER IMPLEMENTATION OF GFAVO IN THE 1990s FOR SO<sub>2</sub> EMISSION REDUCTIONS

### ADDITIONAL FGD APPLICATIONS

Since completion of retrofitting of FGD systems at the large industrial-boiler plant size-range, 35 to 110 MW(e), is not deadlined in Germany until 1993, less than approximately thirty FGD systems have thus far been purchased for such sources, quantified in Table VIII. These FGD-system installations to date, small compared to those at typical utility plants, include many types and in most cases are demonstration plants, installations required on an early schedule due to special provincial requirements or were deadlined for 1988 as part of large plants aggregating more than 110 MW(e). FGD commitments to date thus give no clear verification of the FGD processes that will be preferred and broadly chosen for the retrofits, perhaps more than 200, that will be required to be begun in the next several years. However, current and future operation of comparatively small SO<sub>2</sub> control systems purchased in Europe to date can be expected to be a useful guide in assessment of U.S. industrial boiler application of both the waste throwaway and byproduct recovery type FGD alternatives. Note that at boiler size range, 18 to 35 MW(e), there is presently no control requirement other than to limit SO<sub>2</sub> emissions to 2000 mg/Nm<sup>3</sup> (approximately 1.6 lb per million Btu boiler heat input) and thus compliance simply through use of available fuels e.g. natural gas or low sulfur oil, that can meet this criterion would be expected to be the most logical and inexpensive control means in many instances.

### THE WET LIME/LIMESTONE SCRUBBING APPROACH

Six initial, comparatively small, lime or limestone scrubbing systems are being installed (17) to serve aggregated groups of boilers, each as small as 40 MW(e), or individual small boilers in the size range of 70 to 90 MW(e). These installations parallel activity at electric utility plants where lime/limestone scrubbing is the principal FGD method being used. The very large market for this technology (approximately 90% of all FGD capacity sold in West Germany) has been shared by six principal system suppliers serving West Germany representing a mix of Japanese, U.S., and West German design technologies (17). A typical and simplest wet FGD system arrangement that has been extensively applied in European electric-utility applications (Table V) and could capture a major portion of the market for large industrial boilers incorporates the following features similar to those of systems installed in the U.S. in the 1980s:

- Lime or limestone reagent use, with limestone generally preferred except where a lime supply is deemed preferable so as to ensure good byproduct gypsum color/quality. Note that some limestone sources will impart a gypsum-brightness reduction to as low as 50%. Dark gypsum is limited to cement and mining mortar use. (Unlike circumstances in U.S.A., ground limestone is readily and economically available in West Germany for use in wet FGD systems for industrial boilers since such supply is the norm for the many utility-boiler wet FGD systems already purchased. As a general practice, limestone reagent supply is ground at the mine-source rather than at the boiler FGD site because of comparatively low coal sulfur levels and short limestone shipping distances).
- Spray-tower-type wet absorber design, rubber lined.



- In-situ forced oxidation facilities installed in the sump of the absorber without use of sulfuric acid reagent for pH depression/adjustment.
- Primary dewatering of gypsum slurry by liquid cyclones, with final dewatering by centrifuge or horizontal vacuum filter.
- Production of a throwaway or usable grade of gypsum.
- Single-loop wet-scrubbing design omitting low-pH prescrubbing that has been used extensively in Japan and in many early West German installations for segregated removal of hydrogen chloride and other minor raw gas components. (If applied to wet bottom boilers gypsum whiteness will be impaired at low load due to high unburned carbon emissions.)

Typical design/operating conditions for these comparatively simple, single-loop wet FGD systems are:

- Electrostatic precipitators generally reduce FGD inlet particulate loading to 0.05 grains/SCF.
- With normally used, high-efficiency mist eliminators of the horizontal gas-flow type, FGD liquid carryover is limited so that FGD solid particulate emission is no more than 0.05 grains/SCF.
- SO<sub>2</sub> removal efficiency is 90 to 95% with absorber liquid/gas flow ratio of 90 or more U.S. gallons per 1000 saturated cubic feet.
- Limestone utilization is as high as 99%.
- % oxidation of SO<sub>2</sub> catch to sulfate is 98-99%.

#### NO<sub>x</sub> EMISSION CONTROL

#### OVERVIEW

The emission limit (200 mg/Nm<sup>3</sup>) set in 1984 for West German boilers and plants larger than 110 MW(e) is well beyond the capabilities of combustion modification techniques for bituminous/hard coal and requires the use of flue gas cleaning processes for NO<sub>x</sub> removal. Despite the potential attractiveness of gas cleaning processes that would simultaneously remove both SO<sub>2</sub> and NO<sub>x</sub>, and because of impact of compliance deadlines, the commercially developed and applied SCR process (for NO<sub>x</sub> removal alone) has been most broadly assessed, adapted, and utilized by the electric utility industry. A late 1988 survey indicates that conventional SCR of high-dust design, (upstream of the air preheater) has been installed on 15,250 MW(e) of dry bottom boiler capacity and 2,950 MW(e) of wet bottom (slag tap) boiler capacity. 10,000 MW(e) of SCR has been installed, cold-side, (downstream of FGD), 7,600 MW(e) on wet bottom boilers, 2,400 MW(e) on dry bottom boilers. SCR uses ammonia feed upstream of a catalytic reactor operating at approximately 300 to 400°C (570 to



750°F). Selective noncatalytic reduction (SNCR) systems, which operate at 800 to 1000°C (1470 to 1830°F), have also been installed on three units aggregating 950 MW(e). All ammonia based NO<sub>x</sub> reduction methods have been applied together with the maximum practical utilization of combustion modifications.

## SCR IMPLEMENTATION STRATEGY

Since enactment of the stringent NO<sub>x</sub> regulations in 1984 there have been major concerns and SCR testing programs (18) in Germany to assess catalyst performance and characteristics beforehand. These tests were performed because of questions as to how applicable the commercial SCR experience on new, base-loaded coal-fired boilers in Japan, albeit extensive, is to existing boilers in West Germany. Most of the concerns have been due to the fact that German boilers retrofitted with SCR have a more variable load pattern (more frequent load swings) than those in Japan, which could be expected to cause thermal shock of the catalyst structure. Extensive high-dust SCR field pilot-scale investigation was carried out because possible unique effects of fly ash from German boilers were of concern. Pilot plant results indicated that catalyst poisoning is a major problem in application of high-dust SCR to wet-bottom boilers with recycle of fly ash. Although carefully avoided in Japanese practice and of concern because of expected excessive ammonia slippage leading to fouling of the boiler air preheater, design NO<sub>x</sub> removal efficiency substantially above 80% has been specified for some of the German retrofit SCR applications.

Most of the catalyst for SCR DeNO<sub>x</sub> equipment is honeycomb type. Several West German firms have developed new types of catalyst more resistant to arsenic and poisoning by trace metals typical for West German coal. Generally, catalyst life guarantees have been two years, but it is still too soon to know whether this is a good yardstick. The catalyst cost is quite significant, typically 50% or more of the total cost of the SCR system. Space velocity requirements that have been quoted for West German SCR DeNO<sub>x</sub> systems vary considerably. Space velocity (hr<sup>-1</sup>) is the inverse of the actual flue gas residence time inside the honeycomb channels expressed in hours. A space velocity of 3600 for example would provide a flue gas residence time of 1.0 second in the honeycomb catalyst channels. A very low space velocity number such as 2000 means a higher residence time and a higher degree of performance, with a correspondingly higher cost. Many systems have been quoted with space velocities between 3000 and 6000 in high-dust environment (upstream from the ESP). Uhde-Lentjes has been the only supplier to date willing to quote much higher space velocities (above 6000) but only for cold-side SCR systems (downstream from the wet scrubber). To some extent, a lower space velocity is required for the high-dust case (upstream from the ESP) so that ammonia slip in the passing of the flue gas through the catalyst can be minimized. (More catalyst equates to less slip.) The ammonia slip, if it exceeds about 1 to 2 ppm, can contaminate the fly ash, which is a critical concern in West Germany because substantial fly ash odor can mean that the fly ash cannot be sold or used to manufacture byproduct materials. An excessive amount of flue gas SO<sub>3</sub> and ammonia slip will combine to create very significant air heater fouling. Ammonia slip is not a problem for cold-side SCR systems i.e. those located downstream from the wet scrubber. These latter SCR systems typically involve the following equipment: (a) flue gas reheater wheel, (b) natural gas-fired duct burner to heat the flue gas to 650-700°F, (c) ammonia injection system, (d) honeycomb catalyst layers, and (e) heat recovery in the flue gas reheat wheel to minimize natural gas consumption. Although such cold-side SCR systems cost more, the expected catalyst life is much longer and development and use of high reactivity catalyst will ultimately reduce design operating temperatures. Additionally, catalyst pricing has recently been sharply reduced, adding to the attractiveness of SCR technology.

## LIGNITE FIRED UTILITY BOILERS

Only one lignite fired boiler (approximately 400 MW(e)) has been equipped with SCR (high dust). Approximately 12,200 MW of lignite-fired generating capacity operated by RWE (the largest West German utility company) have been retrofitted with combustion controls and low NO<sub>x</sub> burners to achieve very low NO<sub>x</sub> emission levels. Because lignite, with lower heating value and higher moisture content, requires a larger furnace volume than bituminous coal the modification of combustion controls for NO<sub>x</sub> reduction have been quite effective, and most of the RWE lignite-fired boilers so retrofitted operate at between 175 and 225 ppm NO<sub>x</sub> emission level. RWE has reportedly spent between \$15/kw and \$25/kw for installation of these low NO<sub>x</sub> combustion control systems, versus an average of \$100/kw for SCR and is expected to meet the 100 ppm (200 mg/Nm<sup>3</sup>) NO<sub>x</sub> requirements by an innovative means other than SCR.

## LARGE INDUSTRIAL BOILERS

In view of the ample time to meet deadlines for industrial boiler retrofitting at sizes less than 110 MW(e) there is major incentive for achieving/developing adequate means of NO<sub>x</sub> control for these boilers in all cases without use of selective catalytic reduction. Because of comparatively low capital cost, combustion-modification type retrofit measures are expected to continue to be routinely used so far as practical in the meeting of the NO<sub>x</sub> emission standards for industrial boiler plants smaller than the approximately 110 MW(e) size level. In some or many instances, however, additional reduction in NO<sub>x</sub> emissions beyond that reasonably achievable with retrofitted combustion modifications will be needed in bituminous coal service to meet the emission standard. The national pollution control agency, Umweltbundesamt, has noted that when retrofitting low NO<sub>x</sub> combustion systems, burnout problems increase with decreasing boiler capacity below 100 MW(t), i.e. 37 MW(e), for coal and residual oil fueled plants. A new technology, akin to SNCR, and licensed from EPRI that injects aqueous urea and enhancing additive into the combustion gas stream has been successfully demonstrated in 1987 on a 55 MW(e) coal-fired utility boiler in Sweden (19). 70% NO<sub>x</sub> removal is achieved with no more than 10 ppm of ammonia stack discharge (ammonia slip). Unless flue gas SO<sub>2</sub> level is extremely low, however, this level of ammonia concentration can be expected to adversely affect air preheater reliability.

## WET SIMULTANEOUS SO<sub>2</sub>/NO<sub>x</sub> REMOVAL

It may be speculated that any increment of shortfall of NO<sub>x</sub> removal by combustion modifications may (in the case of boilers in the 35 to 110 MW(e) size range, which are required to retrofit SO<sub>2</sub> removal by 1993), possibly be gained through a modest amount of NO<sub>x</sub> removal achieved simultaneously with SO<sub>2</sub> removal in the FGD operation. In West Germany this would be expected to introduce a very significant new waste management problem because of the need to address contamination of FGD liquid and/or solid waste by soluble nitrogen waste matter. Two wet process-mode alternatives are hypothetically available: absorption-reduction in which NO, the predominant flue-gas NO<sub>x</sub> species, is chemically reduced by ferrous/sulfite reagent with the use of chelate additives and sufficient flue-gas residence time; and absorption-oxidation, using means of converting flue-gas NO to NO<sub>2</sub>, which is more readily absorbed in wet scrubbers than NO. An 800 MW(e) utility installation of absorption-reduction technology in West Germany has failed to meet NO<sub>x</sub> requirements and has been replaced by selective catalytic reduction technology. Its principal shortcoming, it is understood, was the inability to manage FGD nitrogen wastes of the type and amount generated in the scrubbers. Particularly in view of the generally low fuel sulfur-level, i.e. less than 1.5%,

and the direct relationship between rate of FGD NO<sub>2</sub> removal and the magnitude of SO<sub>2</sub>/NO<sub>x</sub> ratio in raw flue gas, absorption-oxidation technology is not readily applied in low-sulfur coal service and has seen no use on coal fired boilers in Europe.

## CONTROL TECHNOLOGIES FOR SMALL INDUSTRIAL BOILERS

A detailed industry survey in West Germany (20) indicates that FGD system suppliers are proposing an array of processes for coal-fired industrial boilers in the range of 1/3 to 18 MW(e), (4 to 170 MM Btu/hr heat input) to provide compliance with the West German 1986 Clean Air Code (TA Luft). These include spray-dryer-type dry scrubbers utilizing dolomitic lime, high calcium lime or sodium-base alkali; various methods of contacting the flue gas with hydrated lime; wet scrubbing with sodium-liquor; wet lime scrubbing yielding byproduct gypsum; as well as a number of other techniques. At the same time, it is expected that many of the smaller boilers will be retired or switched to natural gas or distillate oil fuel. However, significant FGD application on boilers under 18 MW(e) has occurred; a wet lime scrubbing FGD process has been commercially applied in which the thickened scrubber waste solids are processed by being used as the wet feed to an upstream spray-dryer scrubber to yield a dry waste product while avoiding generation of a liquid FGD effluent discharge(21). In some other zero-effluent applications scrubber waste solids are dewatered mechanically to a cake surface moisture content as low as 25%. Ten industrial installations of this proprietary wet FGD system with an aggregate capacity of over 100 MW(e) are in operation in Europe.

## OPERATING EXPERIENCE

### FGD OVERVIEW

Many FGD facilities that have gone into service in West Germany in the mid 1980s are the result of a comparatively modest control program that began prior to 1983 under the jurisdiction of individual provincial governments, which led to 20 FGD applications. Thus, although widespread application of FGD in West Germany is comparatively recent, as of the end of 1984, almost 3000 MW(e) of lime/limestone scrubbing systems, all low sulfur, were in operation, all of single-module slip stream design meeting specific requirements of pre-1983 legislation. These FGD systems installations are reported to have provided acceptable reliability and performance. However, German electric utility specialists note that, with only partial scrubbing of plant flue gas output, these earliest installations were not subjected to typical full-scale electric-utility-industry operating conditions requiring high availability at sustained high/variable load. Additional FGD capacity of more than 30,000 MW(e) originating from the 1983 legislation has now come on-line, primarily wet lime/limestone scrubbing, but a great deal of this capacity is in an early stage of sustained operation without a lengthy period of experience. However, system availability of 90 to 95% is being achieved (on those in service for several years) after 12 months of initial operation. Substantial debugging problems/efforts have typically occurred during the initial operations, primarily with gypsum solids processing equipment i.e., centrifuges, thickeners, filters, dryers, storage facilities, and solids loading and unloading. Operating with characteristically low or moderate coal sulfur level, and applying commercially demonstrated and conservatively designed forced-oxidation systems, no critical technical problems have been experienced or forecast at the present time. Moreover, because of the comparatively low coal sulfur level combined with use of SO<sub>2</sub> removal efficiency in excess of 90%, availability concerns have not required the



use of spare scrubbing modules. Feasibility of regenerative heat exchangers have been effectively demonstrated. FGD retrofitting has not presented insurmountable problems.

#### FGD PROCESS FAILURES

Along with the rapid pace of development, there have been some significant disappointments along the way. One example is a contract potentially tied to 915 MW of wet scrubbing for boilers 14, 15 and 18 of Grosskraftwerke Mannheim (GKM) that was awarded to Walther & Co. The Walther process utilized an ammonia-based wet scrubbing system, which provides very high levels of  $\text{SO}_2$  removal along with optional  $\text{NO}_x$  removal.

The  $\text{SO}_2/\text{NO}_x$  removal performance of the Walther process was verified in pilot plant testing at the GKM site, and the first large FGD system (approximately 240 MW) was installed. The Walther process included several special features, such as two-stage scrubbing at different pH levels, to avoid process problems with byproduct salts and to allow load-following control capability. The byproduct from the Walther process was ammonium fertilizers, which were to have been sold in Europe. Unfortunately, the significant partial pressure of ammonia and the formation of some  $\text{HNO}_3$  mist combined to create an unacceptable stack opacity problem. A technical solution was found in mist eliminator use to mitigate the fume emission. A U.S. plastic mesh design by Kimre used substantial gas pressure drop to alleviate the opacity but contract deadlines were missed by Walther and the ammonia approach at GKM has been replaced by limestone scrubbing.

Another setback was experienced by Davy McKee, with its Wellman Lord regenerable, potassium salt-based process at the 350 MW Buschhaus plant of BKB. Serious reliability problems were encountered due to emission duct and reheater corrosion problems. In addition, oxidation of  $\text{SO}_2$ -catch to sulfate is approximately double that for which the waste sulfate liquor management system was designed and the power plant capacity is presently severely curtailed.

Another system which was declared a failure is the  $\text{DeNO}_x$  capability by wet absorption supplied by Saarberg-Hoelter-Lurgi (SHL) on an 800 MW(e) coal-fired unit at the Heyden IV plant of Pruessenelektra. The SHL  $\text{DeNO}_x$  process was to have provided 85%  $\text{NO}_x$  removal, using a ferrous EDTA complex in the wet limestone scrubbing system. As of January 1, 1988, Dr. Hermann Kraemer, Chairman of the Board of Directors for Pruessenelektra, announced that the Heyden IV plant would be shut down until an SCR  $\text{DeNO}_x$  system could be installed and started up prior to the end of 1988. It is understood that the collected  $\text{NO}_x$  waste processing facility was the performance bottleneck rather than  $\text{NO}_x$  removal capability.

#### SELECTIVE CATALYTIC REDUCTION

In addition to rapid deterioration of catalyst in high dust service on wet bottom boilers recycling fly ash, a number of generic problems in high-dust SCR service have been encountered. These primary operating and/or unresolved issues found in these applications (22) are:

- Initial difficulty in matching the  $\text{NH}_3$  injection pattern to the non-uniform flow of  $\text{NO}_x$  throughout the flow cross-section in the ductwork ahead of the SCR reactor.
- Inability to optimize the  $\text{NH}_3$  injection rate by feedback control of slip  $\text{NH}_3$  due to the unavailability of a proven, reliable  $\text{NH}_3$  monitor.



- Erosion and plugging on systems retrofitted to boilers that fire high ash coal.
- Process control under load swings.
- Spent catalyst disposal.

## PRINCIPAL TECHNOLOGICAL ADVANCEMENTS

Notable commercial developments in Europe that may be significant value in stack gas cleaning applications in the U.S. include a few isolated, high sulfur coal applications, regenerative reheat, FGD wastewater treatment and cold-side SCR.

### HIGH SULFUR COAL APPLICATIONS

European high sulfur coal applications of both the spray dryer and circ. fluid bed scrubber extend the electric utility use of these semidry FGD processes to coal sulfur levels substantially above those of present U.S. installations. The circulating fluid bed scrubber had significant European operating experience in such service, and a substantial basis for broader use of semi-dry scrubbing in the U.S. is developing.

### REGENERATIVE REHEAT

Energy economics in West Germany has provided strong motivation for commercial application of regenerative reheat means in high capacity-factor applications. Rotary air heaters, similar to those commonly used for regenerative combustion-air preheating in large boilers, have been extensively used for this purpose with more than 40 installed. Reliable operation of these heat exchangers has followed from application of suitable materials of construction, favorable continuous performance of electrostatic precipitators dedusting the hot inlet gas, and use of high-efficiency, horizontal-gas-flow mist eliminators limiting carryover of FGD scrubbing slurry. On-line, intermittent, high-pressure water washing has effectively prevented exchanger gas pressure drop (the sum of pressure drop across both sides) from exceeding a nominal 125% of the design value for clean conditions.

Due to the inherent design of the rotary exchanger, 3-5 % of design gas flow leaks from the high-pressure (dirty gas) side to the low-pressure (clean gas) side. Typically, positioning the FGD booster fan on the hot dirty-gas side substantially decreases SO<sub>2</sub> removal. However, a low-leakage design has been developed for this system arrangement that has commercially demonstrated a leakage rate of no more than 0.5% (23).

A comparatively simple, tubular-heat-exchanger type, regenerative reheat system requiring no added flue gas ductwork is also gaining commercial use in Germany after long-term field testing in a lignite FGD installation and may have applicability in the U.S. Tubes exposed to scrubbed gas are supplied in non-metallic construction and circulating water is the heat transfer medium. Four such systems of an aggregate FGD capacity of 1,200 MW(e) are in place.

At one power plant in West Germany, the wet FGD system is located inside a hyperbolic, natural draft cooling tower serving the plant. The FGD outlet gas intermixes with the cooling tower air at a flowrate ratio (cooling tower air/FGD outlet gas) of about 25:1. Because good atmospheric dispersion is ensured by the discharge and mixing of the FGD exhaust with this substantial air flow, the FGD

system is under these circumstance exempted from normal FGD stack exit temperature regulations. For corrosion control a double coating of epoxy is applied to the internal surface of the concrete casing of the cooling tower. The capital cost is reported to be only 20% of that which would be required for a rotary heat exchanger system supplying regenerative reheat.

#### FGD WASTEWATER TREATMENT

The application of usable-by-product-forming, wet FGD systems, common in Germany, results in a significant discharge of brackish/saline wastewater. With typically high chloride levels (0.1 to 0.2%) in German bituminous coal, FGD wastewater management is a substantial problem as would be the case at comparable inland sites in the U.S. producing usable by-product. When discharge to receiving streams is not prohibited, the rate of FGD liquor outfall is usually limited to 95 l/m (25 U.S. gpm) per 100 MW(e). When such saline liquor discharge is allowed, trace metal concentrations, dissolved sulfates, and suspended solids must be reduced to specified levels.

Through wastewater purge treatment carried out in the presence of gypsum precipitation from  $\text{CaSO}_4$ -supersaturated liquid, very low trace metal concentration is achieved prior to outfall discharge (24). A typical wastewater treatment system to accomplish this includes two stages of neutralization, flocculation and sedimentation: the first stage operates at pH 9-10 to remove components such as cadmium and manganese, and the second stage operates at pH 8.5 to remove other components such as zinc and aluminum.

#### COLD-SIDE SCR

Many SCR installations that will commence operations by 1990 are being applied downstream of FGD to limit catalyst poisoning, improve unit availability and, to some extent, because of inadequate plot space to permit normal tie-in of the SCR reactor upstream of the preheater. This activity in Germany may be expected to provide experience needed to gain successful application of SCR in high-sulfur coal service and to optimize design of SCR facilities for especially difficult applications such as wet-bottom boilers, (particularly where it is efficiently coordinated with the installation of FGD).

#### COSTS AND GUARANTEES

The majority of the systems in West Germany are low-sulfur and very successful. Actual removal levels are typically 90% to 97%, the L/G ratios typically exceed 100 gal/MACF, flue gas residence time in the scrubber generally exceeds 10 seconds and the flue gas volumetric flowrates (1400 ACFM per MW(e) at 130°F, saturated) are typically much smaller than those in U.S. wet scrubbing installations. The small flue gas volumes are due to (a) use of regenerative flue gas reheat systems extracting flue gas heat upstream of the scrubber, (b) higher-efficiency steam cycles in West German powerplants, (c) lower excess oxygen and less air preheater leakage in West German powerplants, and (d) systematic differences in air-usage requirements for stoichiometric combustion. The installed cost of a retrofit wet limestone scrubber, typically about \$175/kw in West Germany (per survey of VDEW, Association of German Power Plant Operators, for approximately 400 MW(e) size, with 1 Deutsche Mark = \$U.S. 0.50) would therefore be much higher in the U.S. from the standpoint of differences in flue gas volume alone. (Per a cost study of the Federal Environmental Agency, Umweltbundesamt (25), this actual German retrofit cost is approximately 38% greater than the mid-range of capital cost estimates for FGD for new power plant units.) As might be readily expected

for low-sulfur scrubbers with high SO<sub>2</sub> removal efficiency, spare scrubbers are not utilized or needed in West German installations.

While valid comparisons of retrofit control costs cannot be readily made between German low-sulfur service and U.S. high-sulfur service, an overview of spending for West Germany's mammoth acid precipitation control program may be useful. So far the electric utility industry in West Germany has installed or ordered retrofit gas cleaning systems for control of SO<sub>2</sub> and NO<sub>x</sub> from boiler capacity that presently totals more than 42,000 MW(e), with an equivalent capital cost of about \$10.0 billion broken down as follows:

Wet Flue Gas Desulfurization (FGD)  
38,000 MW at average \$175/kw = \$6.7 billion  
Spray Dryers and Dry FGD  
5,200 MW at average \$75/kw = \$0.4 billion  
Selective Catalytic Reduction (SCR)  
29,000 MW at average \$100/kw = \$2.9 billion

Total to Date: \$10.0 billion

Thus, the investment in retrofit flue gas cleaning equipment in the West German power industry will have amounted to a total of about \$10.0 billion, over a period of five (5) years, from 1984 to first compliance deadlines in 1989. In general the foregoing financial investment in West Germany was basically decided in 1983 with the new GFAVO regulation together with a 1984 governmental accord on NO<sub>x</sub> control. The GFAVO regulation was taken seriously by the entire power industry, and the basic attitude adopted was to proceed immediately with the best and most cost effective solution(s). This attitude still prevails.

Typical performance guarantee levels for West German coal (normally 0.9% to 1.2% sulfur content) are 95% SO<sub>2</sub> removal, emission levels down to less than 100 ppm, and up to 85% DeNO<sub>x</sub> performance to provide compliance with the 100 ppm outlet NO<sub>x</sub> requirement. Combined with high-efficiency electrostatic precipitators, this means that by the 1989 compliance deadlines, coal-fired plants in West Germany (larger than 110 MW) will be producing emissions which are typically equal to or less than burning No. 2 fuel oil. This is in some cases a substantial challenge since some of the boilers fire ballast coal (middlings from coal cleaning) that contains in excess of 30% ash and elevated sulfur levels and with relatively high nitrogen content. To utilize such low-grade fuel it is often fired in specially designed slagging (slag tap) boilers that have inherently high NO<sub>x</sub> emission-forming tendencies. Significant operating experience with controls has been gained only on one such installation and with significant deterioration of SCR catalyst in a typical high-dust mode of operation. Regulations in West Germany also require that powerplant solid wastes be converted into salable products. Therefore, powerplants presently manufacture paving stones and building materials from bottom ash and scrubber sludge, and work is in progress in the gypsum industry to apply processes for manufacturing wallboard plaster, cement, and stucco mortar material to be used in coal mining.

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TABLE I. SUMMARY OF INDUSTRIAL BOILER NSPS (U.S.A.)

| <u>Pollutant/Fuel/Boiler</u>              | <u>% Removal</u> | <u>Lb/million Btu Heat Input</u> |
|-------------------------------------------|------------------|----------------------------------|
| <u>SO<sub>2</sub></u>                     |                  |                                  |
| Coal                                      | 90               | 1.2                              |
| Oil                                       | 90               | 0.8                              |
| Emerging Tech. (Coal)                     | 50               | 0.6                              |
| Emerging Tech. (Oil)                      | 50               | 0.4                              |
| <u>NO<sub>x</sub> (as NO<sub>2</sub>)</u> |                  |                                  |
| Coal, Pulverized                          | -                | 0.7                              |
| Coal, Spreader Stoker                     | -                | 0.6                              |
| Coal, Other Stokers                       | -                | 0.5                              |
| Oil, Synthetic                            | -                | 0.5                              |
| Oil, Resid. (Nit. over 0.35%)             | -                | 0.4                              |
| Oil, Resid. (Nit. up to 0.35%)            | -                | 0.3                              |
| Oil, Distillate                           | -                | 0.2                              |
| Gas                                       | -                | 0.2                              |
| <u>Solid Particulate Matter</u>           |                  |                                  |
| Coal                                      | -                | 0.05                             |
| Oil                                       | -                | 0.10                             |
| Waste                                     | -                | 0.10                             |
| Mixed Fossil + Waste                      | -                | 0.10                             |

TABLE II. ALLOWABLE COAL-FIRED SO<sub>2</sub> EMISSION

| <u>Plant (W.G.),<br/>Unit (U.S.A.), Above<br/>MW(e) Size</u> | <u>Regulatory SO<sub>2</sub> Limit, Lb/MM Btu</u> |                             |                           |
|--------------------------------------------------------------|---------------------------------------------------|-----------------------------|---------------------------|
|                                                              | <u>West Germany</u>                               |                             | <u>U.S.A.</u>             |
|                                                              | <u>New</u>                                        | <u>Existing</u>             | <u>New</u>                |
| 110                                                          | 0.3 (and<br>85% rem.)                             | 0.3 (and<br>85% rem.)       | 1.2 (and<br>70-90%* rem.) |
| 35                                                           | 1.6 (and<br>60% rem.)                             | 1.6 (and<br>60% rem.), 1993 | Do.                       |
| 27                                                           | 1.6                                               | 1.6, 1993                   | Do.                       |
| 18                                                           | Do.                                               | Do.                         | 1.2 (and<br>90% rem.)     |
| 11                                                           | 0.8<br>(approx.)                                  | 1.6, 1989<br>to 1994**      | Do.                       |
| 1/3                                                          | Do.                                               | Do.                         | 1.2 (pending)             |

\* As low as 70% for low sulfur coal

\*\* Plus future 50% retrofit reduction

TABLE III. REQUIREMENTS OF AMENDED EUROPEAN COMMUNITY DIRECTIVE

| <u>Proposed<br/>Directive, 1983</u> | <u>NO<sub>x</sub> Emission Limit, mg NO<sub>2</sub>/Nm<sup>3</sup></u> |     |     |
|-------------------------------------|------------------------------------------------------------------------|-----|-----|
|                                     | Coal                                                                   | Oil | Gas |
| All Plants over<br>50 MW(t):*       |                                                                        |     |     |
| to 1995:                            | 800**                                                                  | 450 | 350 |
| after 1995:                         | 400***                                                                 | 220 | 180 |
| <u>Amended<br/>Directive, 1985</u>  |                                                                        |     |     |
| to 1995:                            |                                                                        |     |     |
| over 50 MW(t)                       |                                                                        | 450 | 350 |
| 50-100 MW(t)                        | 800**                                                                  |     |     |
| 100-300 MW(t)                       | 800                                                                    |     |     |
| over 300 MW(t)                      | 650                                                                    |     |     |
| after 1995:                         |                                                                        |     |     |
| over 50 MW(t)                       |                                                                        | 150 | 100 |
| 50-100 MW(t)                        | 400***                                                                 |     |     |
| over 100 MW(t)                      | 200                                                                    |     |     |

\* 100 MW(t) equals approximately 37 MW(e)

\*\* 1300 mg/Nm<sup>3</sup> for slag tap (wet bottom) furnaces

\*\*\* 800 mg/Nm<sup>3</sup> for slag tap furnaces

TABLE IV. FORECAST OF WEST GERMAN FGD GYPSUM USE

| <u>Market</u>         | <u>Replacement of<br/>Natural Materials, %</u> | <u>Consumption,<br/>MM Met. Ton/Year</u> |
|-----------------------|------------------------------------------------|------------------------------------------|
| Plaster,<br>Wallboard | almost 100                                     | 2.5 - 3                                  |
| Floor<br>Underlayment | 50                                             | 0.6 - 1                                  |
| Portland<br>Cement    | 100                                            | 0.5 - 0.6                                |
| Mining<br>Mortar      | 50                                             | 0.6 - 0.7                                |
| Gypsum/Wood Chipboard | 15                                             | 0.5 - 0.6                                |
| Total                 | -                                              | 5.3 (Avg. Basis)                         |

TABLE V. USABLE GYPSUM PRODUCING, SINGLE-LOOP, IN-SITU  
FORCED OXIDATION FGD

| <u>Station/Unit</u>                           | <u>MW(e)</u>      | <u>Design SO<sub>2</sub> Inlet</u><br><u>(Approx.) mg/Nm<sup>3</sup> (ppmv)</u> | <u>Initial Startup</u> | <u>Reagent</u> |
|-----------------------------------------------|-------------------|---------------------------------------------------------------------------------|------------------------|----------------|
| <u>Deutsche Babcock Supplied: 4,935 MW(e)</u> |                   |                                                                                 |                        |                |
| EVS,<br>Heilbronn,<br>Block 7                 | 2 x 360           | 2,800 (1,000)                                                                   | 1985                   | Limestone      |
| Bremen, Hafen                                 | 300               | 1,800 (650)                                                                     | 1986                   | Limestone      |
| Preag,<br>Staudinger                          | 300               | 2,800 (1,000)                                                                   | 1987                   | Limestone      |
| Bremen, Hafen                                 | 140               | 1,800 (650)                                                                     | 1987                   | Limestone      |
| EVS,<br>Heilbronn,<br>Blocks 3 to 6           | 475               | 2,800 (1,000)                                                                   | 1987                   | Limestone      |
| Steag, Herne                                  | 600               | 2,800 (1,000)                                                                   | 1987                   | Limestone      |
| Steag, Lunen                                  | 500               | 2,800 (1,000)                                                                   | 1987                   | Limestone      |
| Steag,<br>West Voerde                         | 700               | 1,800 (650)                                                                     | 1987                   | Limestone      |
| Steag, Walsum                                 | 560               | 2,800 (1,000)                                                                   | 1988                   | Limestone      |
| VEW Westfalen,<br>Bl. A,B & C                 | 640               | 1,800 (650)                                                                     | 1988                   | Limestone      |
| <u>Steinmueller Supplied: 3,021 MW(e)</u>     |                   |                                                                                 |                        |                |
| Steag/VEW<br>Bergkamen A<br>Module 2          | 2 @<br>0.45 x 747 | 2,400 (850)                                                                     | 1985                   | Lime           |
| RWE/Steag,<br>Voerde, A, B<br>Module 2        | 2 @<br>0.63 x 707 | 1,800 (650)                                                                     | 1985                   | Lime           |
| VEW (Werne)<br>Gersteinwerk K<br>Module 2     | 0.25 x 750        | 1,950 (700)                                                                     | 1987                   | Lime           |
| HEW,<br>Hamburg-Hafen                         | 84                | 2,500 (900)                                                                     | 1987                   | Limestone      |
| NEW,<br>Hamburg-Wedel                         | 261               | 2,800 (1,000)                                                                   | 1987                   | Limestone      |
| BEWAG,<br>Berlin-Oberhavel                    | 200               | 2,800 (1,000)                                                                   | 1988                   | Limestone      |



TABLE V (continued)

| <u>Station/Unit</u>                                         | <u>MW(e)</u>         | <u>Design SO<sub>2</sub> Inlet</u><br><u>(Approx.), mg/Nm<sup>3</sup> (ppmv)</u> | <u>Initial Startup</u> | <u>Reagent</u> |
|-------------------------------------------------------------|----------------------|----------------------------------------------------------------------------------|------------------------|----------------|
| BEWAG<br>Berlin                                             | 225                  | 2,500 (900)                                                                      | 1989                   | Limestone      |
| Steag,<br>Herne 4                                           | 500                  | 2,800 (1,000)                                                                    | 1989                   | Limestone      |
| <u>General Electric Supplied: 1,200 MW(e)</u>               |                      |                                                                                  |                        |                |
| PNEM<br>Amercentrale 8<br>(The Netherlands)                 | 600                  | 2,800 (1,000)                                                                    | 1988                   | Limestone      |
| PGEM<br>Gelderland-13<br>(The Netherlands)                  | 2 x 300              | 2,800 (1,000)                                                                    | 1987,9                 | Limestone      |
| <u>Bischoff Supplied: 4,755 MW(e)</u>                       |                      |                                                                                  |                        |                |
| NWK,<br>Wilhelmshaven<br>Module No 2                        | 450                  | 3,000 (1,080)                                                                    | 1982                   | Lime           |
| Module No 1A                                                | 375                  | 3,000 (1,080)                                                                    | 1985                   | Lime           |
| Isar Amperwerke,<br>Zolling, Bl. 5                          | 450                  | 2,800 (1,000)                                                                    | 1985                   | Limestone      |
| NWK, Kiel                                                   | 350                  | 3,000 (1,080)                                                                    | 1987                   | Lime           |
| NWK, Bremen-<br>Farge                                       | 350                  | 3,000 (1,080)                                                                    | 1988                   | Lime           |
| PZEM,<br>Borssele 11-12<br>(The Netherlands)                | 400                  | 3,500 (1,260)                                                                    | 1987                   | Limestone      |
| GEB,<br>Maasvlakte 1,2<br>Rotterdam<br>(The Netherlands)    | 2 x 540              | 3,500 (1,260)                                                                    | 1987,8                 | Limestone      |
| Elektromark<br>Hagen<br>Werdohl-Elverlingsen<br>E 3 and E 4 | 500                  | 3,340 (1,200)                                                                    | 1988                   | Limestone      |
| Preussen-Elektra<br>KW Staudinger                           | 2 x 250              | 3,000 (1,080)                                                                    | 1988                   | Limestone      |
| TEK<br>Ankara (Turkey)                                      | 2 x 150<br>(lignite) | 21,400 (7,700)                                                                   | 1990                   | Limestone      |

TABLE VI. DRY-SCRUBBING FGD SYSTEMS ON COAL-FIRED BOILERS IN EUROPE

| <u>Plant Name</u>                  | <u>FGD Size, MW(e)</u> | <u>Type System</u> | <u>Design SO<sub>2</sub> Rem.</u> | <u>FGD Supplier</u> | <u>Initial Startup</u> |
|------------------------------------|------------------------|--------------------|-----------------------------------|---------------------|------------------------|
| <u>West Germany</u>                |                        |                    |                                   |                     |                        |
| HKW Ffm West                       | 2 x 75                 | Spray Dryer        | 125 mg/m <sup>3</sup> out         | Lurgi               | 1988/9                 |
| Schwandorf BAG Munich              | 50                     | CFBA               | 95%                               | Lurgi               | 1984                   |
| Borken PREAG-Hanover               | 125                    | CFBA               | 97%                               | Lurgi               | 1987                   |
| Siersdorf EBV-Herzogenrath         | 2 x 100                | CFBA               | 93%                               | Lurgi               | 1986                   |
| Gemeinschafts-Kraftwerke Hannover  | 2 x 130                | Spray Dryer        | 85%                               | Niro                | 1989                   |
| Stadtwerke Erlangen                | 27                     | Spray Dryer        | 85%                               | Niro                | 1987                   |
| Neckarwerke Walheim 1 and 2        | 255                    | Spray Dryer        | 95%                               | Niro                | 1987                   |
| Stadtwerke Braunschweig            | 55                     | Spray Dryer        | 95%                               | Niro                | 1986                   |
| VW Kraftwerk Wolfsburg             | 2 x 130                | Spray Dryer        | 85%                               | Niro                | 1987                   |
| KW Ensdorf Bl. I, Saarland         | 110                    | Spray Dryer        | 95%                               | Niro                | 1987                   |
| KW Ensdorf Bl. III, Saarland       | 300                    | Spray Dryer        | 95%                               | Niro                | 1987                   |
| Huels AG KW Marl                   | 207                    | Spray Dryer        | 93%                               | Niro                | 1988                   |
| KW Wehrden Boiler 5 Völklingen     | 100                    | Spray Dryer        | 60%                               | Flakt               | 1986                   |
| HW Sandreuth Boilers 1-3 Nuremburg | 3 x 39                 | Spray Dryer        | 85%                               | Flakt               | 1987                   |
| Univ. of Freiburg                  | 36                     | Spray Dryer        | 85%                               | Flakt               | 1986                   |

TABLE VI (continued)

| <u>Plant Name</u>                                        | <u>FGD Size, MW(e)</u> | <u>Type System</u> | <u>Design SO<sub>2</sub> Rem.</u> | <u>FGD Supplier</u>    | <u>Initial Startup</u> |
|----------------------------------------------------------|------------------------|--------------------|-----------------------------------|------------------------|------------------------|
| Elektromark<br>Cunokraftwerk<br>Herdecke Bl. 2           | 96                     | Spray<br>Dryer     | 90%                               | Flakt                  | 1987                   |
| KW Mainz-Wiesbaden<br>KWI/Bl. II and III                 | 2 x 100                | Spray<br>Dryer     | 90%                               | Flakt                  | 1988                   |
| KW Münster 1 and 2<br>Stuttgart (oil-fired)              | 181                    | Spray<br>Dryer     | 95%                               | Flakt                  | 1988                   |
| Stadtwerke<br>Duesseldorf,<br>HKW Garath                 | 20                     | Spray<br>Dryer     | -                                 | City of<br>Duesseldorf | 1984                   |
| Stadtwerke<br>Duesseldorf,<br>KW Lausard A-D             | 507                    | Spray<br>Dryer     | -                                 | City of<br>Duesseldorf | 1987/8                 |
| Stadtwerke<br>Duesseldorf,<br>KW Flingern I              | 290                    | Spray<br>Dryer     | -                                 | City of<br>Duesseldorf | 1987                   |
| Stadtwerke<br>Frankfurt,<br>HKW West K5                  | 57                     | Spray<br>Dryer     | -                                 | Unidentified           | 1990                   |
| Stadtwerke<br>Wuerzburg,<br>HKW Friedensbruecke<br>2 & 3 | 2 x 22                 | Spray<br>Dryer     | -                                 | Unidentified           | 1986                   |
| <u>Austria</u>                                           |                        |                    |                                   |                        |                        |
| VKG<br>Duernrohr                                         | 405                    | Spray<br>Dryer     | 90%                               | Niro                   | 1985                   |
| NEWAG<br>Duernrohr                                       | 350                    | Spray<br>Dryer     | 90%                               | Niro                   | 1986                   |
| Salzburger<br>Stadtwerke                                 | 30                     | Spray<br>Dryer     | 97%                               | Niro                   | 1987                   |
| <u>Denmark</u>                                           |                        |                    |                                   |                        |                        |
| Midtkraft el.<br>Studstrupvaerket<br>Bl. III             | 350                    | Spray<br>Dryer     | 92%                               | Niro                   | 1989                   |

TABLE VI (continued)

| <u>Plant Name</u>                           | <u>FGD Size, MW(e)</u> | <u>Type System</u>    | <u>Design SO<sub>2</sub> Rem.</u> | <u>FGD Supplier</u> | <u>Initial Startup</u> |
|---------------------------------------------|------------------------|-----------------------|-----------------------------------|---------------------|------------------------|
| Midtkraft el.<br>Studstrupvaerket<br>Bl. IV | 350                    | Spray<br>Dryer        | 92%                               | Niro                | 1990                   |
| <u>Sweden</u>                               |                        |                       |                                   |                     |                        |
| Aktiebolaget<br>Aroskraft,<br>Västeraas     | 139                    | Spray<br>Dryer        | 80%                               | Niro                | 1987                   |
| Södertälje                                  | 3 x 44                 | Spray<br>Dryer        | 85%                               | Flakt               | 1982                   |
| Sydskraft/<br>Öresundswerket,<br>Malmö      | 55                     | Spray<br>Dryer        | 82%                               | Flakt               | 1983                   |
| KVV No. 1<br>Linköping                      | 29                     | Spray<br>Dryer        | 89%                               | Flakt               | 1985                   |
| Sävenäs, HP3<br>Gothenburg                  | 44                     | Spray<br>Dryer        | 85%                               | Flakt               | 1985                   |
| Västhamsverket<br>Helsingborg               | 60                     | Spray<br>Dryer        | 90%                               | Flakt               | 1986                   |
| <u>Finland</u>                              |                        |                       |                                   |                     |                        |
| Salmisaari<br>Helsinki                      | 210                    | Spray<br>Dryer        | 88.5%                             | Flakt               | 1987                   |
| Imatran Voima<br>Inkoo P.S.                 | 125                    | Humidifier<br>Chamber | 80%                               | Tampella            | 1986                   |
| <u>Italy</u>                                |                        |                       |                                   |                     |                        |
| AEM<br>Brescia                              | 55                     | Spray<br>Dryer        | 81.2%                             | Flakt/Niro          | 1987                   |



TABLE VII. COMPARISON OF LARGEST FGD-USING COUNTRIES

|                                      | <u>West Germany</u> | <u>Japan</u>           | <u>U.S.A.</u> |
|--------------------------------------|---------------------|------------------------|---------------|
| Total Electric Utility FGD,<br>MW(e) | 38,000              | 39,000<br>(Incl. Ind.) | 56,000        |
| Wet FGD, %                           | 90                  | 98                     | 92            |
| Throwaway Waste<br>FGD, %            | 8                   | 3                      | 86            |
| Usable-Gypsum<br>FGD, %              | 87                  | 70                     | 10            |

TABLE VIII. WEST GERMAN FOSSIL-FIRED  
PLANTS, 18 TO 110 MW(e)

| <u>Fueling</u>            | <u>Number of Boilers</u> | <u>Aggregate MW(e)</u> |
|---------------------------|--------------------------|------------------------|
| Bituminous,<br>Dry Bottom | 69                       | 2,900                  |
| Bituminous,<br>Wet Bottom | 186                      | 9,860                  |
| Bituminous,<br>Balance    | 26                       | 675                    |
| Lignite                   | 81                       | 4,250                  |
| Residual Oil              | <u>249</u>               | <u>9,515</u>           |
|                           | 611                      | 27,200                 |

## FLUE GAS CLEANING IN POWER STATIONS IN THE NETHERLANDS

by: ir. F.W. van der Bruggen

KEMA

Arnhem, The Netherlands

ir. J.M. Koppius-Odink

EPON

Zwolle, The Netherlands

### ABSTRACT

The growing concern about the influence of air pollution on human health during the seventies and the effects of acid precipitation during the past decade led to the formulation of a more stringent air pollution policy. The objectives of this policy, the resulting standards for stationary sources and the role of the electric power supply companies in air pollution abatement will be described.

One of the measures taken by the electricity supply companies was the introduction of flue gas desulphurization. The criteria that led to the sole application of the lime/limestone - gypsum technology will be described and a short overview will be given of the systems that have been installed.

Finally, the experience with the first operational installation will be given in more detail: desulphurization efficiency, availability, costs, and the behaviour in the installation of several other compounds such as HCl, HF, particulate material and heavy metals.

### AIR POLLUTION POLICY IN THE NETHERLANDS

#### OBJECTIVES OF THE DUTCH AIR POLLUTION POLICY

Air pollution by sulphur dioxide and nitrogen oxides has been a focus of public interest in the Netherlands for many years. Initially this interest was generated by fear of the threat to human health. At present the major concern is caused by the consequences of acid deposition. The objective of the current air pollution policy is to maintain a good ambient air quality and to substantially reduce acid deposition.

Acid deposition in the Netherlands is attributed to three compounds: SO<sub>2</sub>, NO<sub>x</sub> and NH<sub>3</sub>, the latter being released by the intensive bioindustry. In the year 1980, the average acid deposition amounted to 5,800 equivalent of potential acid per hectare. The Dutch policy, formulated in 1984, is based on a reduction to 3,000 equivalent of potential acid per hectare in the year 2000. With this objective in mind and taking into account the technical and economic possibilities the following emission reductions were derived with 1980 as the reference year: SO<sub>2</sub> 70%, NO<sub>x</sub> 33% and NH<sub>3</sub> 50%.

At present the government is evaluating the results of the anti-acidification policy. In an interim report published in December 1987 it was concluded that the 3000 eq.H<sup>+</sup> per hectare deposition target will not bring sufficient relief to ecosystems like the woods, heather and fens. Tentative target values between 900 and 2100 eq. H<sup>+</sup> per hectare were mentioned. The consequences for the Dutch air pollution policy are not yet clear.

It must be borne in mind that all deposition values include a substantial import from abroad.

#### STATUTORY INSTRUMENTS

The most important statutory instrument is the Air Pollution Act of 1970. The provisions of this general law are worked out in detail in General Administrative Orders. Considering only stationary sources, the most important administrative orders issued until now are:

- O The plant order (1972), governing the plants that are subject to licence under the terms of the air pollution law.
- O The order on the sulphur content of fuel (1974): this governs the maximum sulphur content of mineral oil, gaseous and solid fuels. The values are regularly updated.
- O The order on ambient air quality with respect to sulphur dioxide and black smoke (1986), specifying the permitted and target concentrations of sulphur dioxide and black smoke in the ambient air.
- O The order on ambient air quality with respect to nitrogen dioxide (1987), specifying the permitted and target concentrations of nitrogen dioxide in ambient air.
- O The order on emission limits for combustion plants (1987). Emission, limits of all coal fired plants and of gas- or oil fired plants from 2,5 MW<sub>th</sub> and upwards are established for sulphur dioxide, nitrogen oxides and dust. In many cases the values of this ordinance are based on the "Großfeuerungsanlagen Verordnung" in the Federal Republic of Germany. The most important emission limits of this ordinance are summarized in Table 1.

Natural gas plays an important role in the energy household of the Netherlands. Gas turbines and gas engines are widely used and special NO limits have been formulated in the administrative order for such installations. Generally speaking after 1-1-1990 the emission limit is 200 g/GJ NO<sub>2</sub> for a single gasturbine and 135 g/GJ NO<sub>2</sub> for gas turbine installations like combined cycle units.

Finally there are special regulations for refineries. International aspects play a predominant role in this case. The limits for sulphur dioxide are specified as follows:

- O 1986 2500 mg/m<sup>3</sup> (furnace emissions only)
- O 1991 2000 mg/m<sup>3</sup> (including process emissions)
- O 1996 1500 mg/m<sup>3</sup> (including process emissions).



## MEASURES TAKEN BY THE ELECTRICITY SUPPLY COMPANIES IN THE NETHERLANDS

### ELECTRICITY PRODUCTION IN THE NETHERLANDS

Table 2 shows a number of key-data concerning electricity supply in the Netherlands in 1987. Coal and natural gas play a predominant role at this moment and this will last over the next decade. Two new 600 MWe coal fired units have been commissioned: Amer-9 at Geertruidenberg to be onstream in July 1993 and one year later Hemweg-8 in Amsterdam.

Recently, Sep, the Dutch Electricity Generating Board announced the erection of another 600 MWe coal fired unit: Maasvlakte - 3 in Rotterdam (July 1997) as well as two 600 MWe natural gas fired units at Eemshaven (July 1995 and 1996). Gas for these units will be imported from Norway and is contracted at coal parity. Furthermore power, equivalent with 1400 MWe generating capacity has been contracted in France and the FRG and will be imported. Sep expects that until 1995 the generating capacity of smaller scale cogeneration units will expend with 1000 MWe. Until 1996 4270 MWe of old generating capacity, of which 1000 MWe is coal fired, will be decommissioned.

All these plans must secure the reliability of the electricity supply in the Netherlands, where the consumption is expected to grow at a rate of 1.9% per year until 1998.

Finally, one of the most striking facts in the new plans of Sep is the decision to build an Integrated Coal Gasification Combined Cycle System of between 200 and 300 MWe at Maascentrale. This technology has been in the focus of interest of the electric supply companies during the last decade. Several projects have been under way but were abandoned. Growing awareness of the potential environmental benefits led to the decision to build this demonstration unit. The unit must be on stream in 1993. Sep commissioned the engineering firm Comprimo to perform a detailed study of three gasification processes: Texaco, Shell, British Gas/Lurgi, so that by the end of this year the gasification process can be selected.

The new 600 MWe Maasvlakte -3 unit mentioned above could be the first big scale commercial IGCC power station. This is of course dependent of the success of the demonstration plant.

### EMISSIONS OF SULPHUR DIOXIDE BY THE POWER STATIONS

Almost entirely as a result of the energy policy of the Dutch government, the total amount of sulphur dioxide emitted by power stations showed substantial fluctuations over the past decade. By 1975 the total emission figure had fallen to 30,000 t/a due to large-scale use of domestic natural gas. The energy policy promulgated in 1974 after the first energy crisis, reserved this natural gas predominantly for smaller scale applications such as heating and industry and forced the power companies to change over again to heavy fuel oil. This resulted in an increase of SO<sub>2</sub> emissions up to 189,000 t/a in 1980. Since 1980 more natural gas was made available again to the power companies: by 1986 emissions had fallen to 66,000 t/a.

By installing FGD in existing and new coal fired power plants, and by shutting down 1000 MWe of old uncontrolled coal fired generating capacity the emissions will be reduced below 40,000 t/a by 1993 and will stabilize at this level until the year 1997. The further development of SO<sub>2</sub>-emissions will depend on the government decision on the extension of nuclear generating capacity and on the technical success of integrated coal gasification combined cycle systems.

#### EMISSIONS OF NITROGEN OXIDES BY THE POWER STATIONS

The current yearly NO<sub>x</sub> emission is approximately 80.000 t and will stabilize at this level until 1995, in spite of the start-up of new coal fired generating capacity. As for SO<sub>2</sub>, the development of the NO<sub>x</sub>-emission level is dependent on the application of nuclear energy.

The standards for NO<sub>x</sub> in the administrative order on emission limits for combustion plants are based on the application of so called primary measures, i.e. combustion and burner modifications. Flue gas cleaning by selective catalytic reduction by NH<sub>3</sub> is kept in a reserve in case of disappointing results of the primary measures or in case of further tightening of the emissions standards.

The electricity supply companies have set up a costly concerted NO<sub>x</sub> abatement programme to evaluate the possibilities of several primary NO<sub>x</sub> measures:

- A demonstration of HTNR burners and two stage combustion for front-wall coal-fired boilers (170 MWe, target value 400 mg/m<sup>3</sup> NO<sub>2</sub>).
- A demonstration of "In-furnace NO<sub>x</sub> reduction" in a natural gas/oil-fired boiler (180 MWe).
- Evaluation of furnace and burner modifications in three tangentially fired coal boilers that have been converted from oil/gas to coal/gas firing.
- Evaluation of the effect of repowering 10 existing natural-gas-fired units with a gas turbine. The exhaust gas from the gas turbine is used as combustion air for the boiler. Besides an increase in generating efficiency, a marked decrease in NO<sub>x</sub> emission is expected and has in fact been realized.

#### CONTRIBUTION OF THE POWER STATIONS IN THE TOTAL DUTCH EMISSIONS

The Ministry of the Environment published actual and estimated values for the emissions of SO<sub>2</sub> and NO<sub>x</sub> in the years 1980, 1985 and 2000. The figures are summarized in Table 3.

The actual values of 1980 and 1985 show a marked decrease of the SO<sub>2</sub> emission, both national and by the power stations. Furthermore the share in SO<sub>2</sub> emissions by the power stations decreased. The figures for NO<sub>x</sub> hardly show any change.

Looking at the estimated values for 2000 it will be obvious that the primary goals of the Air pollution policy, reductions of 70% for SO<sub>2</sub> and 33% for NO<sub>x</sub> will not be realized. Fortunately the estimated values are too pessimistic. They are based on coal and coal/nuclear scenario's that have been superseded by the recently published plans of the Sep described before. At this moment the expectation is that the SO<sub>2</sub>- and NO<sub>x</sub>-emissions by power stations will amount to 25,000 t/a and 60,000 t respectively in the year 2000.

It can be concluded that the power supply companies in the Netherlands contribute substantially to the reduction of total national SO<sub>2</sub> emissions. The NO<sub>x</sub> emissions, that were already relatively low in 1980, will slowly decrease to 60.000 t/a, being less than 15% of the total national emission.

## FLUE GAS DESULFURIZATION IN POWER STATIONS IN THE NETHERLANDS

### CURRENT SITUATION

In 1978 the Dutch power supply companies were confronted with FGD for the first time. One of the power companies, EPON, was obliged to study the possibility of installing an FGD-unit for 20% of the flue gas from their new 600 MWe coal-fired unit Gelderland-13. The intention was to demonstrate FGD technology under circumstances typical for Dutch power stations, which are load following and have regular changes in coal properties. By 1980, before completion of this study, the power company was obliged to build an FGD installation for 50% of the flue gas from Gelderland-13. This installation has been in operation since April 1985. The limestone-gypsum technology was selected. To ensure a good gypsum quality, a prescubber was integrated in the installation. Experiences with this installation will be described in more detail in the next chapter.

In 1980, at the request of the provincial authorities, another power company, EPZ, started a study to investigate the technical possibilities and economic consequences of FGD for their 645 MWe Amer-8 power station. At the special request of the government the study first concentrated on non-gypsum producing systems, but finally resulted in the decision to build an in-situ-forced-oxidation limestone/gypsum system for 50% of the flue gas-flow. As a result of the growing awareness of the "acid-rain problem", the government policy tightened during the last stage of engineering of this 50% installation, resulting in the obligation to build a unit for the total flue gas flow. Operation started in March 1988.

During the same period it was decided to convert three natural gas/oil fired blocks into natural gas/coal-firing. These units, Borssele-12, Maasvlakte 1 and 2 had to be equipped with desulphurization for the total flue gas flow. The lime/gypsum system was selected for Borssele-12, the two others use limestone. The installations have all gone on stream.

The two new 600 MWe coal-fired blocks that have already been commissioned, will both be equipped with lime/limestone gypsum technology. Maasvlakte-3 will use either IGCC or a pulverized coal-fired boiler with FGD. As mentioned, this decision will be based on the results of the IGCC demonstration at Maascentrale.



## SELECTION CRITERIA FOR AN FGD SYSTEM IN THE NETHERLANDS

The selection of an FGD system by a power company in the Netherlands is based on the following criteria:

- O Reliability of the technology.
- O Environmentally acceptable applicability of the sulphur containing byproduct.
- O Availability of base chemicals.
- O Desulphurization efficiency.
- O Secondary pollution problems such as waste water, noise or visual nuisance.
- O Safety related problems.
- O Costs.

According to law operation of a power station without its FGD is only allowed 240 hours per year with a maximum uninterrupted period of 72 hours. In the present situation coal-fired power stations in the Netherlands are used for base-load because of the low generating costs. This means that a high reliability for the unit as a whole, and consequently for the FGD installation is necessary.

The best way to get a proper insight into the technological status of an emerging technology is to have discussions with manufacturers and to visit operating plants. The acquired information can be translated to the local situation. Several of these studies showed over and over that during the period of decisionmaking the penetration of the lime/limestone scrubbing technology either with sludge (USA) or gypsum (FRG, Japan) as a byproduct, was steadily increasing worldwide.

Process design is mature, mechanical problems are solved, the whole system is simplified. This results in high reliability in the FRG and Japan and increasing reliability in the USA in low sulphur coal applications. Other technologies could only point to a limited number of references. Regenerative systems are just too complicated, the Walther process showed serious shortcomings. The only technology that showed some promise was spray-drying absorption and this technology has been seriously considered for the Amer-8 power station.

But here is another major selection factor: the usefulness of the byproduct. In the Dutch situation it is absolutely necessary under the current circumstances to have environmentally sound applications. Possible byproducts are:  $\text{CaSO}_4$ - $\text{CaSO}_3$ -sludge, gypsum, ammonium sulphate, sulphur, sulphuric acid and SDA-byproduct. Sludge producing systems are out of the question in the Netherlands. Extensive research has been carried out and is still in progress to develop bulk applications for the SDA-byproduct. Very promising in this respect is application in sand-limebrick, a widely used building material in the Netherlands. Unfortunately, despite these efforts, there is still no secured outlet for the product other than disposal, which makes the very promising spray drying absorption technology unacceptable up till now.

Consumption of gypsum and related products in the Netherlands is about 600,000 t/a, and because there are no domestic natural resources, until recently the total amount had to be imported (Table 4). The quality of FGD gypsum is such, that it can replace natural gypsum in most of its traditional applications. It is reasonable to assume that FGD gypsum from Dutch power stations can at least partly replace these imports. Developments of new building materials like self-levelling anhydrite-based floors and hollow building blocks can increase the market potential. However, it must be clear that FGD activities abroad, especially in the FRG, will influence the Dutch market, and that a gypsum surplus could arise by the year 2000 if all planned coal fired power stations are equipped with gypsum producing FGD processes.

Ammonium sulphate is a byproduct of several industrial processes and is produced in amounts of about 4,000,000 t/a in Western Europe. Despite this, the fertilizer industry shows interest in ammonium sulphate from FGD systems.

Sulphur is readily marketable in the Netherlands. The national consumption cannot be covered by the domestic production capacity of approximately 250 000 t. Yearly imports fluctuate around 300 000 t, with the fertilizer industry as the main consumer, for the production of sulphuric acid.

It is unlikely that sulphuric acid produced by FGD systems can penetrate into the fertilizer industry, so that the current market is more or less limited to the foreign trade deficit of about 250,000 t/a.

Availability of the base chemicals is no problem. Domestic limestone resources are limited, but are very extended at a reasonable distance in neighbouring countries. Cheap bulk transportation along waterways is possible.

A major secondary environmental problem is the waste water from wet scrubbing systems. Licensing authorities lay strong emphasis on suspended solids and heavy metals. A very efficient system for removal of heavy metals from FGD waste water, based on co-precipitation of hydroxides and sulphides, has been developed and is applied successfully. The target values, formulated by the licensing authorities and shown in Table 5, can be reached with this technology. Sludge from these waste water treatment installations, the main constituent being gypsum, is mixed with the coal feed to the boiler and "fired".

With a proper design noise and visual problems can be prevented.

Lime and limestone systems do not involve special safety aspects. The ammonium sulphate system introduces the problem of bulk ammonia storage, where special precautions have to be taken. Safety problems also arise in regenerative systems with sulphur production where  $H_2S$  is an intermediate product.

In general, wet scrubbers can easily reach desulphurization efficiencies of 90%. It must be borne in mind however that the desulphurization efficiency of the total system can be influenced by the reheat system. The often applied rotary - type heat - exchangers can have an internal leakage of 5%. The efficiency of spray-drying-absorption is somewhat limited. High efficiencies can only be reached at the cost of a high reagent -  $SO_2$  ratio.

Especially for non-lime/limestone systems reliable cost estimates are very problematic. Budget proposals by manufacturers tend to be too optimistic, and the costs that have to be made by the utility itself are often underestimated. Comparison of rough cost estimates for the Dutch situation reveal the ranking shown in Table 6, with the lime/limestone - gypsum system as the base case. The lime(stone)/gypsum, Walther and spray-drying absorption process do not differ substantially in specific costs per kWh; however the Wellman Lord process is more expensive.

Looking at all these factors, it will be obvious that up till now lime/limestone gypsum systems have been selected by the Dutch power supply companies: high reliability, acceptable costs, good desulphurization efficiency, sales potential for the byproduct, no safety problems and secondary environmental problems that can easily be mastered. Reliability of the ammonium sulphate process is insufficient, the Wellman-Lord process is comparatively complex and expensive, the SDA-process produces byproduct that has so far proved unsaleable.

#### SHORT DESCRIPTION OF THE FGD SYSTEMS IN THE NETHERLANDS

Table 7 shows a general overview of the five FGD installations built in the Netherlands. Gelderland-13 was built in two 50% stages. The first stage was equipped with a prescrubber and separate oxidation vessels, but has meanwhile been adapted to the in-situ forced-oxidation mode without prescrubbing. The second stage which will be operational by December 1988 will also use this technology.

Operational experience of most units is only limited, but no serious problems were encountered during the start-up phase. Major point of concern is the gypsum quality, especially colour, crystal size and shape. The two latter factors strongly influence the dewatering properties of the filter cake on a belt filter or in a horizontal continuous centrifuge. It proved to be difficult to reach the required moisture content of 10% by weight.

The colour or "whiteness" of the gypsum could be improved by removal of brown mud from the scrubbing liquid, either by a hydroclone system or by increasing the upward flow in the gypsum settling tank. The desired chloride content of less than 100  $\mu\text{g/g}$  is realized in all installations, also the one that uses sea water for make-up (Borssele-12).

All installations have spraytowers, the biggest one treating 1,925,000  $\text{m}^3/\text{h}$  (STP) of flue gas, equivalent of 645 MWe (Amer-8).

In a number of scrubbers damage of the the rubber lining was found, which probably has to be attributed to erosion by the washing suspension.

## OPERATIONAL EXPERIENCE WITH THE GELDERLAND-13 FGD INSTALLATION

### DEMONSTRATION PROGRAMME

The Gelderland-13 FGD installation was the first operational system in the Netherlands. The government subsidized part of the investment cost and in exchange required to perform a two-year demonstration program comprising of:

- O Recording of operational parameters such as desulphurization efficiency, availability, gypsum quality, waste water quality.
- O Recording of maintenance items, and costs.
- O Total operating costs.
- O Special measurements to study behaviour of other flue gas compounds like HCl, HF and particulates and of the heavy metals As, Cd, Cr, Cu, Ni, Hg, Pb, Zn and Se.

The demonstration programme was also subsidized by the government, and performed under the auspices of NOVEM BV, the Netherlands Agency for Energy and Environment.

### DESCRIPTION OF THE PLANT

Figure 1 shows a simplified flow scheme of the installation. Flue gas enters the rotary-type heat exchanger with a temperature of approximately 120°C and is cooled down to 80°C. In the prescrubber acid compounds such as HCl, HF and SO<sub>3</sub>, as well as part of the fly-ash, are removed. Moreover, the flue gas is saturated with water vapour and cooled down to approximately 48°C.

The prescrubber is a cylindrical vessel with 2 spray layers. The circulating liquid is acid water, pH < 1. Part of this water is continuously blown off to the waste water treatment, to prevent accumulation of compounds removed from the flue-gas. This waste water loss and the evaporation losses are replenished with water from the river Rhine.

The flue gas passes one package of a two-pass open-louver-type demister and enters the scrubber, where it is contacted with a slurry mainly consisting of calcium sulphite and- sulphate. The scrubber is a cylindrical vessel with 4 spray layers, including a spare one. Desired efficiency can normally be reached by using 3 spray layers.

The cleaned flue gas leaves the spray tower through 2 packets of a two-pass open louver type demister, is mixed with about 10% of recirculated reheated gas to evaporate any droplets, reheated in the rotary-type heat exchanger and released through the stack, where it is mixed with the untreated flue gas.

The SO<sub>2</sub> absorption capability of the washing suspension is maintained by adding freshly prepared limestone suspension. Moreover, NaOH is added; the manufacturer claims better growth of the gypsum crystals. Suspension extracted from the scrubber vessel is sent to the two oxidation vessels in series where compressed air is blown into the liquid. The desired pH for oxidation, approximately 4,5 is reached by injection of concentrated H<sub>2</sub>SO<sub>4</sub>. Any unreacted limestone is also neutralized. A small amount of the oxidized suspension is returned to the scrubber as seed crystals.



The gypsum suspension is concentrated in a thickener. The concentrated underflow is the feed for 4 batch-wise operating vertical-axis centrifuges, in which the gypsum is dewatered to below 10% moisture. The thickener overflow is partly used for make up of fresh limestone suspension, the remainder is sent to the waste water treatment. Gypsum is stored in a so-called Eurosilo.

The waste water treatment plant processes two flows: blow-off from the prescrubber and part of the thickener-overflow. Neutralization and further pH adjustment take place in two steps with a lime suspension. After addition of polyelectrolyte, the solids are separated in a thickener. The sludge is dewatered on a belt-filter and sent to the coal storage for "firing". A simplified scheme of the waste water treatment plant is given in Figure 2.

#### MAJOR OPERATIONAL PARAMETERS

The most important operational parameter is the desulphurization efficiency of the complete installation, inclusive of the regenerative rotary type heat exchanger. Figure 3 shows the daily averages of the desulphurization efficiency for the period of January 1986 till July 1987. The efficiency fluctuates between 82% and 97%, with an average value of 89.6%. The SO<sub>2</sub> balance over the same period is given in Table 8. From this balance it is clear that the availability of the FGD installation has a strong influence on the total SO<sub>2</sub> emission during the period considered.

Table 9 gives an overview of the availability of the FGD installation. Major causes of the lengthy outages were:

- O Rapidly developing leakages in the prescrubber vessel after small liner failures.
- O Loosening of the spray nozzles in the prescrubber after practically complete dissolution of the fastening bolts (either titanium grade-12 or stainless steel DIN 14539).
- O Rapidly developing leakage in the scrubber vessel after liner damage due to erosion by the washing liquid.
- O Failure of plastic packets in the regenerative rotary type heat exchanger due a difference in thermal expansion with the steel packets. Pieces of this plastic reached the prescrubbing circulating loop through the flue gas and caused damage to liners of pumps, the prescrubber vessel and liquid lines.

During the period considered 31,530 t of gypsum was produced. The gypsum was sold to the Belgian firm Gyproc for the production of gypsum board. A major problem was maintaining the standard "whiteness" agreed upon with this firm. All other requirements, moisture content and chemical composition, could easily be realized, as is shown in Table 10.

The heavy metal content was determined during the special measuring campaigns that will be described later. The heavy metal content in the gypsum was of the same order as in the limestone.

The removal of heavy metals in the waste water treatment plant functioned very well at a pH of 9. The target values, described by the licensing authorities, could be reached for most heavy metals even without sodium sulphide addition. This is shown in Table 11.

## MAINTENANCE

Maintenance costs during the period under consideration are summarized in Table 12.

Beside the facts already mentioned above, other important maintenance issues were:

- O Leakage of compensators in flue gas ducts.
- O Malfunctioning of flue gas dampers due to fly ash deposits.
- O Wear of rotating seals of pumps and mixers.
- O Deposits on pH electrodes.
- O Wear of the gypsum transportation system.

## COSTS

The investment costs for the installation amounted to NLG 123,000,000 (NLG 410 per kW) based on 1986. The operational costs are summarized in Table 13.

It is clear that capital is by far the most important cost factor in this installation. However it must be kept in mind, that the investment costs are relatively high. The installation has a prescrubber, separate oxidation vessels, and the major part of the gypsum handling system and the waste water treatment plant are designed for the full 630 MWe of the boiler. The power consumption is somewhat above 1 % of the power produced. The proceeds of the gypsum are negligible in the whole cost scene.

## SPECIAL MEASURING CAMPAIGNS

To establish the influence of the installation on other flue gas compounds three special measuring campaigns were performed. The major results of these campaigns are summarized in Table 14.

The influence on nitrogen oxides is negligible, although the washing liquid contains approximately 300 mg/l nitrate. The removal efficiency for HCl is somewhat better than for HF. The bulk of these compounds is already washed out in the prescrubber. As expected, the prescrubber has only minor influence on SO<sub>2</sub>-removal.

The fly ash inlet concentration varied between 25 and 90 mg/m<sup>3</sup> and in all cases the outlet dust concentration was clearly below 10 mg/m<sup>3</sup>. Enrichment of this dust with gypsum as a result of the entrainment of washing liquid, could not be established. This can easily be explained by the excellent performance of the demisters. With a special impingement measuring method developed by TÜV-Hamburg (FRG) it could be established that the content of droplets in the range of 3-100 µm was very low: 9 mg/m<sup>3</sup> at full load and 24 mg/m<sup>3</sup> at 60 % load. The experience of TÜV-Hamburg is that entrained droplets of washing liquid are in this size range.

For two other volatile compounds in the flue gas, Se and Hg, no accurate balances could be made. The determination of the concentrations in the flue gas turned out to be inaccurate. However, from analyses of the blow-off from the

prescrubber and, to a lesser extent from the main scrubbing loop it can be concluded that a substantial amount of these compounds is removed: between 20-60 g/h Se and approximately 2 g/h Hg is transferred to the waste water treatment plant, where Hg is removed for the greatest part. Removal of Se in the waste water treatment plant is still questionable. Concentrations between 380 and 850 µg/l in the effluent were found. Another complication of the presence of Se is the fact that the sludge from the waste water treatment plant contains such an amount of this element that it has to be considered as chemical waste (legal limit 50 g/kg). The same holds for the element fluor. This is found in concentrations of more than 20 mg/g in the sludge, which is the legal limit for chemical waste.

The general picture for several other heavy metals is clear: major sources are the flue gas and the limestone feed; the input through the riverwater supply is negligible. Outlets for heavy metals are the waste water treatment plant and the gypsum; the amount in the cleaned flue gas is negligible. Table 15 gives the amounts of ingoing and outgoing heavy metals in g/h.

In July 1987 the operation of the unit was stopped; the prescrubber and oxidation vessels were dismantled. The oxidation air is now blown into the hold tank of the main scrubber vessel. Operation was restarted in December 1987. Initially problems were encountered in reaching the desired desulphurization efficiency of 90 %.

#### FINAL REMARKS

The environmental and technological measures described are only part of the efforts to maintain a healthy living climate in the densely populated Netherlands. In the context of this conference attention was focussed on air pollution abatement, especially sulphur dioxide where the electric supply companies make an important contribution.

For many other pollutants, measures are in preparation or have already been introduced. The present government objective is to achieve as much as possible an integration of these measures so that an optimum protection of air, water and soil will be realized.

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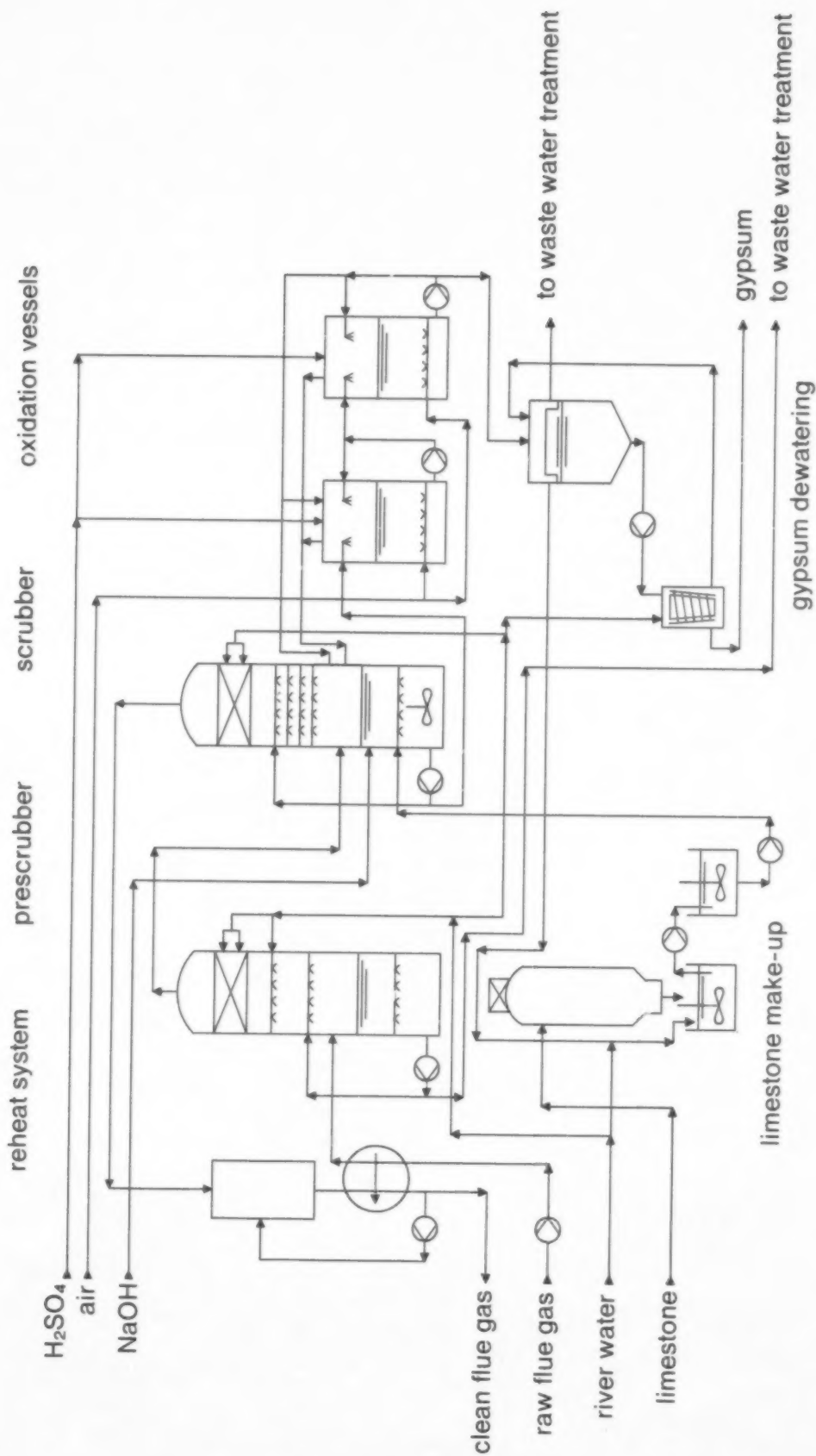


Figure 1 FGD at Gelderland-13 power station



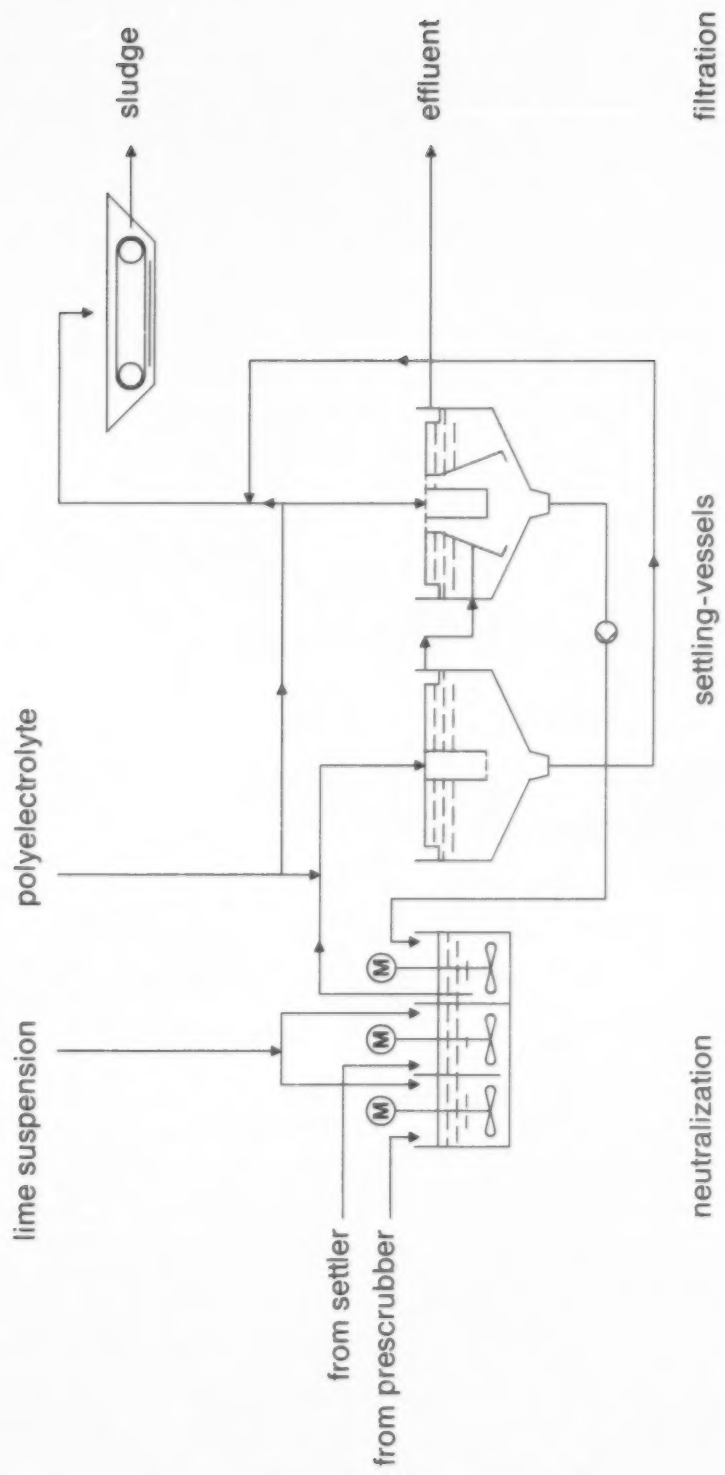


Figure 2 Waste water treatment plant

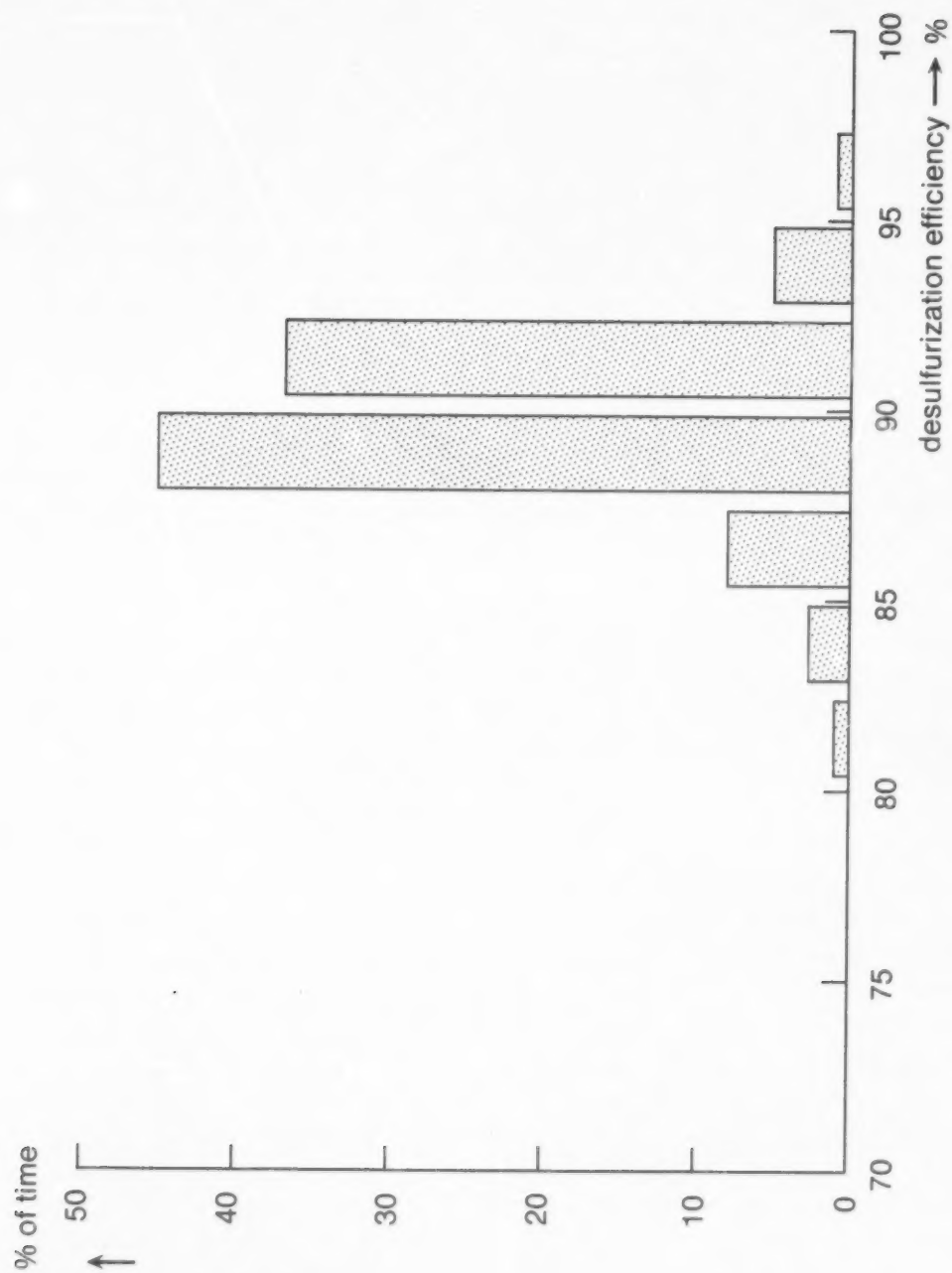


Figure 3 Daily average of desulfurization efficiency

TABLE 1. EMISSION LIMITS FOR COMBUSTION INSTALLATIONS (1987)  
(mg/m<sup>3</sup>, NTP)

|                                                                     | New plant                           | existing plant                                                                          |
|---------------------------------------------------------------------|-------------------------------------|-----------------------------------------------------------------------------------------|
| 2 Sulphur dioxide                                                   |                                     |                                                                                         |
| Coal > 300 MW <sub>th</sub>                                         | 400 + 85%FGD                        | max. 0,8% S in coal<br>or 400 + 85% FGD<br>for units in<br>operation after<br>1-1-1986. |
| < 300 MW <sub>th</sub>                                              | 700                                 | max. 1,2% S in coal                                                                     |
| Oil > 300 MW <sub>th</sub>                                          | 400 + 85%FGD                        | 1700<br>400 + 85% or FGD<br>for units in<br>operation after<br>1-1-1986.                |
| < 300 MW <sub>th</sub>                                              | 1700                                | 1700                                                                                    |
| Nitrogen oxides (as NO <sub>2</sub> )                               |                                     |                                                                                         |
| Coal > 300 MW <sub>th</sub>                                         | 400 <sup>a</sup> - 800 <sup>b</sup> | 1100 (pulverized<br>coal)<br>1000 (others)                                              |
| < 300 MW <sub>th</sub>                                              | 500 <sup>a</sup> - 800 <sup>b</sup> |                                                                                         |
| Oil > 2,5 MW <sub>th</sub>                                          | 300 <sup>a</sup> - 450 <sup>b</sup> | 700                                                                                     |
| Gas > 2,5 MW <sub>th</sub>                                          | 200 <sup>a</sup> - 350 <sup>b</sup> | 500                                                                                     |
| Particulate material.                                               |                                     |                                                                                         |
| Coal                                                                | 50                                  | existing licence                                                                        |
| a) licence after August 1, 1988<br>b) licence before August 1, 1988 |                                     |                                                                                         |

The standards in Table 1 are specified in mg/m<sup>3</sup> (NTP) and are normalized to an oxygen content of 6% for pulverized coal firing, and 3% for gas and oil firing.

TABLE 2. KEY DATA ON ELECTRICITY SUPPLY IN THE NETHERLANDS IN 1987

|                         |        |     |
|-------------------------|--------|-----|
| Generating capacity     | 15,140 | MW  |
| Maximum load            | 10,330 | MW  |
| Net output              | 56.6   | TWh |
| Electricity imports     | 3.64   | TWh |
| Shares of fossile fuels |        |     |
| Coal                    | 30,8%  |     |
| Oil                     | 1,6%   |     |
| Natural gas             | 60,4%  |     |
| blast furnace gas       | 7,2%   |     |

TABLE 3. SO<sub>2</sub> AND NO<sub>x</sub> EMISSIONS IN THE NETHERLANDS (1000 t/a)

|                       | actual<br>1980 | actual<br>1985 | estimated<br>2000<br>coal scenario | estimated<br>2000<br>nuclear scenario |
|-----------------------|----------------|----------------|------------------------------------|---------------------------------------|
| SO <sub>2</sub> total | 477            | 275            | 240                                | 217                                   |
| power stations        | 189            | 65             | 76                                 | 53                                    |
| %                     | 39,6           | 23,6           | 31,6                               | 24,4                                  |
| NO <sub>x</sub> total | 538            | 537            | 559                                | 538                                   |
| power stations        | 89             | 83             | 103                                | 82                                    |
| %                     | 16,5           | 15,4           | 18,4                               | 15,2                                  |

TABLE 4. GYPSUM IMPORTS IN THE NETHERLANDS IN 1985

|                       |           |
|-----------------------|-----------|
| dihydrate + anhydrite | 270,000 t |
| β-hemihydrate         | 56,000 t  |
| Plaster               | 94,000 t  |
| plaster board         | 148,000 t |
| building blocks       | 118,000 t |
|                       | 636,000 t |



TABLE 5. TARGET VALUES FOR HEAVY METAL CONCENTRATIONS IN WASTE WATER  
FROM WET SCRUBBING SYSTEMS ( $\mu\text{g/kg}$ )

|    |    |
|----|----|
| As | 20 |
| Cd | 1  |
| Cr | 15 |
| Cu | 10 |
| Hg | 1  |
| Ni | 15 |
| Pb | 50 |
| Zn | 50 |

TABLE 6. COST COMPARISON OF FGD SYSTEMS

|                               | Investment cost | cost per kWh |
|-------------------------------|-----------------|--------------|
| Limestone gypsum              | 100             | 100          |
| * Ammonium sulphate (Walther) | 100             | 100          |
| Wellman-Lord                  | 115-130         | 120-140      |
| Spray drying absorption       | 85- 90          | 90-100       |

\* Ammonium sulphate solution as byproduct.

TABLE 7. FGD SYSTEMS IN THE NETHERLANDS

| Utility Company            | EPZ                                              | PZEM                         | EZH*                          | EPON**                                         |                                                                   |
|----------------------------|--------------------------------------------------|------------------------------|-------------------------------|------------------------------------------------|-------------------------------------------------------------------|
| Unit                       | Amer 8                                           | Borssele 12                  | Maasvlakte 1-2                | Gelderland-13                                  |                                                                   |
| Location                   | Geertruidenberg                                  | Borssele                     | Rotterdam                     | Nijmegen                                       |                                                                   |
| Boiler size MWe            | 645                                              | 405                          | 517                           | 630                                            |                                                                   |
| FGD size MWe               | 645                                              | 405                          | 517                           | 309                                            | 321                                                               |
| Total efficiency %         | 88                                               | 89                           | 88                            | 90                                             |                                                                   |
| Technology                 | GEESI                                            | Bischoff                     | Bischoff                      | GEESI/<br>Mitsui                               | GEESI                                                             |
| Engineer                   | ESTS                                             | Comprimo                     | Tebodin                       | ESTS                                           | ESTS                                                              |
| Start-up date              | March 88                                         | July 88                      | No 2 July 87<br>No 1 sept 88  | April 85                                       | Dec. 88                                                           |
| Process type               | Limestone/<br>gypsum                             | Lime/gypsum<br>seawater      | Limestone/<br>gypsum          | Lime-<br>stone/<br>gypsum+<br>prescrubber      | Lime-<br>stone/<br>gypsum                                         |
| Scrubber type              | spray tower                                      | spray tower                  | spray tower                   | spray tower                                    | Spray-<br>tower                                                   |
| Number of scrubbers        | 1                                                | 1                            | 2                             | 1                                              |                                                                   |
| scrubber size m            | 38x17                                            | 65x12,2                      | 52x10,5                       | 34x11,2                                        | 33x13,6                                                           |
| L/G L/m <sup>3</sup>       | 11,5                                             | 7,46                         | 9,08                          | 10                                             | -                                                                 |
| number of pumps            | 3 + 1                                            | 3                            | 2 x 3                         | 3 + 1                                          | 3 + 1                                                             |
| gypsum oxidation           | in situ                                          | in situ                      | in situ                       | separate,<br>2 vessels<br>in series            | in situ                                                           |
| gypsum dewatering          | hydrocyclones<br>+ contineous<br>hor. centrifuge | hydrocyclones<br>belt-filter | hydrocyclones+<br>belt-filter | settler+<br>4 vertical<br>batch<br>centrifuges | hydro<br>cyclones<br>3 ver-<br>tical<br>batch<br>centri-<br>fuges |
| demister type              | GEESI open<br>louver<br>1 packet                 | Euroform<br>LTH 2100         | Euroform<br>LTH 2100          | GEESI<br>open<br>louver<br>2 pagkage           | GEESI<br>open<br>Louver<br>1<br>package                           |
| Reheat system              | heat off-gas<br>existing gas-<br>turbine         | rotary-type                  | rotary type                   | rotary-<br>type                                | none<br><br>both<br>flue<br>gas<br>flows<br>will be<br>mixed      |
| Reheat tempera-<br>ture °C | 50 -> 65                                         | 48 -> 92                     | 49 -> 90,5                    | 48 -> 90                                       | -                                                                 |

\* Maasvlakte 1 and 2 have the same design.

\*\* The first stage of the Gelderland-13 FGD has been reconstructed. The prescrubber and separate oxidation vessels have been dismantled; the installation has been in operation according to the in-situ forced-oxidation mode since December 1987.

TABLE 8. SO<sub>2</sub> BALANCE OF GELDERLAND-13 POWER STATION OVER THE PERIOD  
JANUARY 1986 - JULY 1987

|                                                               |          |       |
|---------------------------------------------------------------|----------|-------|
| Amount of SO <sub>2</sub> to FGD                              | 16 150 t | 100%  |
| SO <sub>2</sub> emission through scrubber                     | 1 190 t  | 7.4%  |
| SO <sub>2</sub> emission as a consequence of reheater leakage | 325 t    | 2.0%  |
| SO <sub>2</sub> emission during outages of the FGD            | 1 700 t  | 10.5% |
| SO <sub>2</sub> removed in scrubber                           | 12 935 t | 80.1% |

TABLE 9. AVAILABILITY OF THE GELDERLAND-13 FGD INSTALLATION DURING THE  
PERIOD OF JANUARY 1986 - JULY 1987

|                                      |        |
|--------------------------------------|--------|
| Total hours during period considered | 13,567 |
| Operational hours of FGD             | 11,401 |
| FGD outages                          | 1,804  |
| FGD available during boiler outage   | 362    |
| Availabilitiy                        | 86.1%  |
| Operability                          | 84 %   |

DURATION AND NUMBER OF OUTAGES OF FGD

|        |    |
|--------|----|
| 0- 8 h | 18 |
| 8-50 h | 13 |
| > 50 h | 9  |

TABLE 10. QUALITY OF THE GYPSUM PRODUCED AT GELDERLAND-13 POWER STATION

|                                | Requirement | Average value   | maximum  |
|--------------------------------|-------------|-----------------|----------|
| Fe <sub>2</sub> O <sub>3</sub> | 1.5 %       | 0.14 %          | 0.36 %   |
| Al <sub>2</sub> O <sub>3</sub> |             | 0.24 %          | 0.70 %   |
| SiO <sub>2</sub>               | 1.0 %       | 0.47 %          | 1.24 %   |
| MgO                            | 0.1 %       | 0.045 %         | 0.13 %   |
| K <sub>2</sub> O               | 0.1 %       | 0.037 %         | 0.08 %   |
| Na <sub>2</sub> O              | 0.04 %      | 0.01 %          | 0.02 %   |
| Cl                             | 0.01 %      | 0.002 %         | 0.0045 % |
| CO <sub>3</sub>                |             | 0.16 %          | 1.3 %    |
| SO <sub>2</sub>                | 0.25 %      | 0.13 %          | 1.9 %    |
| Moisture                       | 8 %         |                 |          |
| As                             | -           |                 |          |
| Cd                             |             | 2.0 - 2.5 mg/kg |          |
| Cu                             |             | 0.03 mg/kg      |          |
| Cr                             |             | 1.2 mg/kg       |          |
| Hg                             |             | 1.3 - 1.6 mg/kg |          |
| Ni                             |             | 0.6 mg/kg       |          |
| Pb                             |             | 1.0 - 1.4 mg/kg |          |
| Zn                             |             | 6 - 10 mg/kg    |          |
| Se                             |             | < 5 mg/kg       |          |
|                                |             | < 3 mg/kg       |          |

TABLE 11. HEAVY METAL REMOVAL IN THE WASTE WATER TREATMENT INSTALLATION  
(CONCENTRATIONS IN µg/kg)

|    | target value | effluent pH=7 | effluent pH= 9 |
|----|--------------|---------------|----------------|
| As | 20           | < 3           | < 5            |
| Cd | 1            | < 15          | < 1            |
| Cu | 10           | < 100         | < 10           |
| Cr | 15           | < 15          | < 15           |
| Hg | 1            | < 10          | < 2            |
| Pb | 50           | < 10          | < 10           |
| Ni | 15           | < 200         | < 15           |
| Zn | 50           | < 300         | < 50           |



TABLE 12. MAINTENANCE COSTS OF FGD INSTALLATION OF GELDERLAND-13  
POWER STATION DURING THE PERIOD OF JANUARY 1986 - JULY 1987

|                                      |     |           |        |
|--------------------------------------|-----|-----------|--------|
| Computer                             | NLG | 162,000   | 9.5 %  |
| Preventive, electric and instruments | NLG | 122,000   | 7.1 %  |
| Repair, electric and instruments     | NLG | 392,000   | 22.9 % |
| Preventive, mechanical               | NLG | 214,000   | 12.5 % |
| Repair, mechanical                   | NLG | 823,000   | 48.0 % |
| total                                | NLG | 1,713,000 |        |

TABLE 13. OPERATIONAL AND SPECIFIC COSTS OF GELDERLAND-13 FGD DURING  
THE PERIOD OF JANUARY 1986 - JULY 1987

|                                                  |              |        |
|--------------------------------------------------|--------------|--------|
| Capital (7% interest rate,<br>15 years lifetime) | 20,934,000   | 77.8 % |
| Maintenance                                      | 1,713,000    | 6.4 %  |
| Personel                                         | 1,239,000    | 4.6 %  |
| Power (37,000 MWh, NLG 40/MWh)                   | 1,480,000    | 5.5 %  |
| Limestone (18,300 t, NLG 56/t)                   | 1,024,000 )  |        |
| Sulfuric acid (860 t, NLG 210/t)                 | 180,600 )    |        |
| Sodium hydroxide (966 t, NLG 235/t)              | 227,000 )    | 6 %    |
| Other chemicals                                  | 206,000 )    |        |
| Gypsum 31,500t                                   | - 83,000     | -0.3 % |
| Total                                            | 26,921,400   |        |
| Per ton of SO <sub>2</sub> removed               | : NLG 2.080  |        |
| Per kWh                                          | : NLG 0.0084 |        |
| Per ton of gypsum produced                       | : NLG 854    |        |

TABLE 14. INFLUENCE OF PRESCRUBBER AND TOTAL INSTALLATION ON OTHER  
FLUE-GAS COMPOUNDS

|                 | Removal efficiency (%) |           |
|-----------------|------------------------|-----------|
|                 | Prescrubber            | total FGD |
| SO <sub>2</sub> | 10                     | 90        |
| NO              | -                      | -         |
| NO <sub>2</sub> | -                      | -         |
| HCl             | 90                     | 85-97     |
| HF              | 75-80                  | 75-80     |
| Fly-ash         | 80                     | 85-95     |

TABLE 15. AMOUNTS OF HEAVY METALS IN INGOING AND OUTGOING FLOWS  
(g/h)

|    | Inlet    |           | Outlet      |        |
|----|----------|-----------|-------------|--------|
|    | flue gas | limestone | waste water | gypsum |
| As | 9 - 10   | 4 - 6     | 2           | 6 - 9  |
| Cd | 0.1-0.3  | 0.1-0.2   | 0.2-0.3     | 0.1    |
| Cr | -        | 3         | 3           | 5      |
| Cu | 5 - 11   | 2         | 4 - 5       | 4      |
| Ni | 4 - 19   | 1 - 2     | 5           | 3 - 4  |
| Pb | 5 - 8    | 11 - 17   | 2 - 4       | 16     |
| Zn | 11 - 46  | 8 - 14    | 13 - 17     | < 17   |

RECENT DEVELOPMENTS IN SO<sub>2</sub> AND NO<sub>x</sub> ABATEMENT TECHNOLOGY  
IN JAPAN AND CHINA

by

Junpei Ando  
Chuo University  
Kasuga, Bunkyo-ku, Tokyo, Japan

and

Norman Kaplan  
Air and Energy Engineering Research Laboratory  
U.S. Environmental Protection Agency  
Research Triangle Park, NC 27711

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ABSTRACT

Wet process FGD plants in Japan number about 1500 with a total capacity of about 50,000 MW (150 million Nm<sup>3</sup>/h) and achieve 90-98% SO<sub>2</sub> removal. Limestone-gypsum process, the major process for utility boilers, has been significantly improved to attain better operability with lower cost, resulting in the construction of new plants and the modification of old plants to apply the improved systems.

Dry and semi-dry lime injection is applied at more than 1000 municipal solid waste (MSW) incinerators to reduce SO<sub>2</sub> and HCl emissions by 30-90%. The MSW incinerators emit small amounts of mercury and dioxins but the most harmful methyl mercury and 2,3,7,8 tetrachlorodibenzoparadioxin are not detectable or are present in very low concentrations. Dry lime based sorbent processes have been tested for coal-fired boilers.

More than 250 selective catalytic reduction (SCR) plants are in operation, mainly at utility boilers in Japan to reduce NO<sub>x</sub> emissions by 60-85%. Selective noncatalytic reduction (SNR) is used at about 50 smaller gas sources including MSW incinerators to reduce NO<sub>x</sub> emissions by 20-50%.

Combined systems, to remove both SO<sub>2</sub> and NO<sub>x</sub>, using SCR followed by FGD are popular in Japan and are also applied in the Federal Republic of Germany (FRG). Systems with FGD followed by SCR and simultaneous removal processes are also in commercial use in Japan and FRG.



In China,  $\text{SO}_2$  emissions have increased to nearly 20 million metric tons/year. Photochemical smog problems also have been experienced. Some boilers and furnaces have been equipped with FGD and some nitric acid plants with SCR.

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation.

## INTRODUCTION

The information contained in this paper was obtained primarily from plant management and operators, and from equipment manufacturers or vendors. Some information was obtained from other sources. One of the coauthors (Junpei Ando) has visited many of the facilities, reported in the paper. Very little of the information reported was obtained by direct testing and observation by the authors.

### 1. PROBLEMS WITH $\text{SO}_2$ AND $\text{NO}_x$ IN JAPAN AND CHINA

#### 1.1 STATUS IN JAPAN

The  $\text{SO}_2$  emissions in Japan were reduced from 5 million metric tons in 1967 to less than 1 million metric tons in 1986 (Figure 1), resulting in the decrease in yearly average ambient  $\text{SO}_2$  concentrations from 0.05-0.07 ppm to 0.005-0.01 ppm in large cities and industrial regions. The  $\text{NO}_x$  emissions reached a maximum of about 2 million metric tons in 1974 and have since decreased to 1.2 million metric tons. Annual average ambient  $\text{NO}_x$  concentrations in large cities increased from 0.02-0.03 ppm in 1974 to 0.025-0.035 ppm in 1979 despite the decrease in  $\text{NO}_x$  emissions, and have since decreased slightly. The ambient  $\text{NO}$  concentration, however, continued to decrease between 1974 and 1979.

Dieback of older Japanese cedar trees is observed in a fairly wide region 10-50 km northwest of Tokyo (1). The yearly average rain pH in the region as well as in Tokyo held constant at around 4.5 for the past 10 years. The nitric acid concentration has increased while the sulfuric acid concentration has decreased. The lake waters in the region have shown no tendency to become acidic. Oxidant concentrations (mainly  $\text{O}_3$ ) are high in the region and occasionally exceed 0.12 ppm, the photochemical smog warning level. Although the cause of the dieback is not certain, it may be due to high oxidant concentrations.

#### 1.2 STATUS IN CHINA

The  $\text{SO}_2$  emissions in China have increased to approach 20 million metric tons/year (Figure 1). Coal consumption has exceeded 600 million metric tons/year, accounting for most of the  $\text{SO}_2$  emissions. Since the per capita energy consumption in China is less than 10% that in the U.S.,  $\text{SO}_2$  emissions in China will continue to increase as per capita energy consumption rises, and may eventually exceed U.S. emissions.

In the south central part of China including Chongqing (Figure 2) where they use high sulfur coal containing 3-5% S, the rain pH has fallen to 4.2-4.3. Coal-fired utility boilers under construction in Chongqing will be required to apply FGD (Section 5).

Large cities in northern China including Beijing (Peking) have serious air pollution in winter due to the coal burning for house heating. Coal cleaning and gasification are being studied. Photochemical smog problems have been experienced in Lanzhou where there is extensive industrial activity including oil refining. Efforts to abate  $\text{NO}_x$  emissions have begun.

Currently, the air pollutant of most concern is particulate matter (PM). Sources of PM include: coal-fired boilers, various other industrial sources with insufficient PM control, and fugitive emissions from unpaved roads due to increasing automobile traffic on them. While control of PM is of primary importance, efforts to control other air pollutants including  $\text{SO}_2$  and  $\text{NO}_x$  are expected to increase with time.

## 2. DEVELOPMENTS IN $\text{SO}_2$ AND $\text{NO}_x$ ABATEMENT TECHNOLOGY IN JAPAN

### 2.1 STATUS OF FGD AND $\text{NO}_x$ ABATEMENT

The wet limestone-gypsum FGD process is used to control about 150 large gas sources including most coal-fired boilers. The total controlled capacity is about 28,000 MW, which is about 50% of the total FGD capacity. The process has been significantly improved (Section 2.2). No significant developments have been made with the double alkali type and regenerable processes.

Nearly 100 magnesia scrubbing plants with capacities ranging from 10,000 to 200,000  $\text{Nm}^3/\text{h}$  of gas were recently constructed. These plants produce a waste sulfate liquor, using a magnesium hydroxide slurry (30-35% concentration) as the absorbent. The  $\text{Mg}(\text{OH})_2$  absorbent is obtained from sea water by lime addition followed by thickening of the resulting precipitate. Magnesium hydroxide has also been used to replace sodium hydroxide at many once-through sodium alkali scrubbing plants that produce waste sulfate liquor, since magnesium hydroxide is less than half as expensive as sodium hydroxide.

Dry and semi-dry lime injection processes have not been used for large gas sources because of their low removal efficiency, but are used at many MSW incinerators (Section 4). Dry lime-based sorbent processes for coal-fired boilers have been tested. Two commercial dry activated coke process plants are in operation (Section 3.3). These plants remove  $\text{SO}_2$  and  $\text{NO}_x$  simultaneously by adsorption on coke.

For  $\text{NO}_x$  abatement, more than 100,000 stationary sources have applied combustion modifications including fuel switching from heavy oil to kerosene, two-stage combustion, flue gas recirculation, and low- $\text{NO}_x$  burners to reduce  $\text{NO}_x$  emissions by 20-70%. The  $\text{NO}_x$  emission standards set by the Central Government are based on the advanced technology of combustion modification but actual emissions are controlled by more stringent local regulations. For a new large coal-fired boiler, the  $\text{NO}_x$  emission standard is 200 ppm but  $\text{NO}_x$  emissions are usually reduced to 45-100 ppm by SCR based on local agreements.

Table 1 classifies coal-fired utility boilers in Japan according to flue gas cleaning facilities. As shown, 11 smaller and older boilers apply neither FGD nor SCR and burn domestic low-sulfur coal containing 0.2-0.3% S. Eleven larger boilers have FGD units without SCR. Twenty-six boilers, representing 62% of total capacity, have both FGD and SCR units. The wet limestone-gypsum FGD process and SCR technology developed in Japan have also been used extensively in FRG.

### 2.2 IMPROVEMENTS IN WET LIMESTONE-GYPSUM PROCESS

Major types of the wet limestone-gypsum processes used in Japan are shown in Figure 3 and Table 2. The most popular is type IV that has a prescrubber in a separate liquor loop and an oxidizer. The prescrubber removes chlorides, fluorides, and fly ash that tend to lower the  $\text{SO}_2$  removal efficiency and gypsum purity. The type IV plants, however, are costly at 20,000-30,000 yen (\$160-240)/kW for investment and they consume about 2% of the power generated by the boiler.

Efforts have been made to simplify the type IV process to reduce the cost. MMEC constructed two plants (Minato and Ishikawa, Table 2) with the type VII process without a

prescrubber. This process as well as the type III process uses sodium sulfate as an additive to depress the adverse effect of fluoride. The plants have been operating well, reducing  $\text{SO}_2$  from 950-1200 to 20-40 ppm at a stoichiometry of 1.05 with a power consumption of 1.8-2.0%.

MHI developed process types VIII and IX, both of which have air spargers in the scrubber tanks without separate oxidizers. The oxidation in the scrubber promotes  $\text{SO}_2$  removal by lowering the  $\text{SO}_2$  vapor pressure of the absorbent slurry (2). The type IX has no prescrubber and is the simplest.

MHI recently modified process type IV to types VIII and IX. The power consumption of the modified plants has been 1.6%. A new Kashima Minami FGD plant constructed by MHI with the type IX process started operation in late 1987 on a 146 MW oil-fired boiler (Figure 4). The plant cost 10,000 yen (\$80)/kW and reduces  $\text{SO}_2$  from 1640 to 80 ppm with a low power consumption of about 1%.

MHI is constructing two 700 MW type VIII plants for coal-fired boilers in Japan (Table 2) and type IX plants in China (Section 5). BHK is constructing one type IX plant for a 1000 MW boiler (Table 2). The adverse effect of the fluoride may be depressed by an additive.

The type V process developed by Chiyoda uses the Jet bubbling reactor. The type VI process developed by KHI uses magnesite as the additive. With these processes little or no decrease in  $\text{SO}_2$  removal efficiency may occur when the prescrubber is eliminated. One Chiyoda process plant was constructed in the U.S. Many plants by the KHI process have been constructed in FRG including new ones of the type IX process.

Chubu Electric is constructing three 700 MW coal-fired boilers at a new station at Hekinan. Limestone-gypsum processes by MHI, KHI, and Chiyoda will be used (Table 2). The flue gas will contain about 800 ppm  $\text{SO}_2$ . Design requirements are to reduce  $\text{SO}_2$  emissions below 50 ppm, particulates below 10 mg/Nm<sup>3</sup>, and to maintain gypsum purity above 95%. All FGD plants will have air spargers in the scrubber tanks without separate oxidizers. Prescrubbers will be used for the plants by MHI and Chiyoda but not for the KHI plant. All plants are to have non-leak type (multi-tube type) gas-gas heaters and wet ESPs similarly to the Yokosuka plant of Tokyo Electric constructed by MHI (3).  $\text{NO}_x$  emissions will be reduced below 45 ppm using SCR.

### 2.3 DRY LIME-BASED SORBENT PROCESS

Hokkaido Electric (4) has developed a dry process using an active sorbent composed of lime and fly ash that may be, to some extent, similar to that reported by Jozewicz and co-authors (5). Slaked lime, calcium sulfate, and fly ash from coal are mixed, pelletized with water, and treated at 100°C using steam to prepare porous pellets 3-10 mm in size. The pellets are used as the sorbent in a moving bed to remove about 90% of  $\text{SO}_2$  in a coal-fired flue gas leaving an ESP around 140°C (Figure 5). About 90% of the remaining dust in the gas is also removed.

Following the tests at a 1000 Nm<sup>3</sup>/h pilot plant, a demonstration plant (50,000 Nm<sup>3</sup>/h) has been operated since December 1987. The spent sorbent is used as the source of calcium sulfate. It has been found that  $\text{H}_2\text{O}$  and  $\text{NO}_x$  in the gas promote  $\text{SO}_2$  removal and that the absorbed  $\text{SO}_2$  forms  $\text{CaSO}_3$ .

It seems that the fly ash and calcium sulfate promote  $\text{SO}_2$  absorption mainly by increasing the reactive surface area of lime particles. The details of the process will be given at the present symposium by Hokkaido Electric and Hitachi Ltd.

Another dry process was tested at a small scale using the process described by the schematic in Figure 6. Pulverized limestone was injected into the boiler. The particulate



matter in the gas exiting the boiler, including fly ash and calcium compounds from the limestone, was removed in an ESP. This material was then classified into coarse and fine fractions, above and below 5  $\mu\text{m}$  size, respectively. The coarse fraction, containing mostly fly ash was discarded. The fine fraction, containing fly ash,  $\text{CaO}$ , and  $\text{CaSO}_4$ , was treated with water and steam to obtain a reactive sorbent which was subsequently pulverized to less than 15  $\mu\text{m}$  and used in an absorber vessel at 100-150°C to achieve an overall  $\text{SO}_2$  removal rate in excess of 80%.

Hitachi Zosen has tested the injection of very finely pulverized limestone smaller than 2  $\mu\text{m}$  into a furnace at a 1000  $\text{Nm}^3/\text{h}$  pilot plant. Usually very fine particles are readily agglomerated to become less reactive. Hitachi Zosen has developed a way to attain a good dispersion of the particles, so that 80-85% of the  $\text{SO}_2$  could be removed at a stoichiometry of 2 with a reaction time of 3.3 sec. It should be noted that this performance has only been achieved in a small pilot operation for short periods. A larger pilot plant (10,000  $\text{Nm}^3/\text{h}$ ) is expected to be operational in 1989.

## 2.4 $\text{NO}_x$ REMOVAL TECHNOLOGY

About 250 SCR plants are in operation mainly at utility boilers to reduce  $\text{NO}_x$  emissions by 60-90% (Figure 7). Parallel-flow catalysts with a pitch of about 7 mm are mainly used for dust-rich gases while those with 4-6 mm pitch or pellet catalysts are used for dust-lean gases. The cost of SCR plants including initial charge of catalyst is 6,000-8,000 yen (\$48-64)/kW for coal-, 3000-5000 yen (\$24-40)/kW for oil-, and 2000-3000 yen (\$16-24)/kW for gas-fired boilers. The catalyst life has been 4-5 years for coal, 7-10 years for oil, and more than 10 years for gas.

For MSW incinerator flue gas, an HCl resistant catalyst is used. For wet bottom coal-fired boilers in FRG, arsenic resistant catalysts have been developed.

Selective noncatalytic reduction (SNR) or Thermal De $\text{NO}_x$  has been used at about 50 smaller boilers and furnaces including MSW incinerators to reduce  $\text{NO}_x$  emissions by 20-50% (Figure 7). Recently a urea solution has been sprayed directly into MSW incinerators at around 900°C to reduce  $\text{NO}_x$  emissions by about 30%, using 0.5 mol urea to 1 mol  $\text{NO}_x$ . One mole of urea ( $\text{CO}(\text{NH}_2)_2$ ) may decompose at high temperature to give 2 moles of  $\text{NH}_3$  which react with  $\text{NO}_x$ . The advantage of this process is that urea is easier to store and handle than  $\text{NH}_3$ .

In-furnace  $\text{NO}_x$  reduction (reburning) has been applied at more than 10 boilers mainly by MHI and Hitachi Zosen, and has achieved about 50% additional  $\text{NO}_x$  reduction over levels achieved by combustion modification. The process requires a little larger furnace volume than normal and therefore may not be suitable for existing boilers.

## 2.5 FLUIDIZED BED COMBUSTION (FBC)

About 20 FBC industrial boilers with capacities of 2-23 MW equivalent are in operation burning low-sulfur coal. Most of them use limestone as the fluid bed sorbent and reduce  $\text{SO}_2$  emissions by 50-80%.  $\text{NO}_x$  emissions are usually maintained below about 200 ppm by combustion control.

A 50 MW prototype FBC plant has been operated by EPDC since 1987. More than 90%  $\text{SO}_2$  removal has been achieved while burning coals containing 0.6-0.8% S, with a  $\text{CaO}/\text{SO}_2$  mole ratio of 4-6 in the fluid bed.  $\text{NO}_x$  has been maintained at 167-210 ppm by two-stage combustion. An SCR unit is installed for further  $\text{NO}_x$  reduction when needed.



## 2.6 COST OF $\text{SO}_2$ AND $\text{NO}_x$ REMOVAL

Figure 8 shows the estimated cost of removing 1  $\text{Nm}^3$  of  $\text{SO}_2$  or  $\text{NO}_x$  from flue gas from a 500 MW coal-fired utility boiler. It includes annual capital charges at 15% of the investment. Combustion modification (CM) and the combination of CM with in-furnace  $\text{NO}_x$  reduction are less costly and may reduce  $\text{NO}_x$  emissions by up to about 80% for less than about 50 yen (\$0.40) per  $\text{Nm}^3$  of  $\text{NO}_x$ .

Using SCR to reduce  $\text{NO}_x$  by 80%, or using a combination of SCR and CM to reduce  $\text{NO}_x$  by 90%, the estimated cost to remove 1  $\text{Nm}^3$  of  $\text{NO}_x$  is about 300 yen (\$2.40) (\$1060/metric ton  $\text{NO}_x$ ). Controlling  $\text{SO}_2$  at the 90% level using the improved wet limestone-gypsum processes, the estimated cost to remove 1  $\text{Nm}^3$  of  $\text{SO}_2$  is about 250-300 yen (\$2.00-2.40) (\$635 to \$762/metric ton  $\text{SO}_2$ ).  $\text{SO}_2$  removal costs by dry lime/limestone processes are not certain due to the wide variation of the by-product disposal cost.

## 3. COMBINED AND SIMULTANEOUS $\text{SO}_2$ AND $\text{NO}_x$ REMOVAL

### 3.1 SCR FOLLOWED BY FGD

The combination of high-dust SCR and wet FGD (Figure 9, A) has become popular in Japan and FRG to remove  $\text{SO}_2$  and  $\text{NO}_x$  from flue gas produced by coal combustion. Parallel-flow catalysts resistant to  $\text{SO}_2$  and dust are used for SCR. The low-dust SCR system (Figure 9, B) is also used in Japan. Both systems have generally worked well but occasionally have problems.

The major problem is related to the oxidation of  $\text{SO}_2$  in the SCR reactor. Usually 0.2-2% of the  $\text{SO}_2$  is oxidized to  $\text{SO}_3$ . This has various effects on the flue gas cleaning system. For low sulfur coals,  $\text{SO}_3$  may improve the dust removal efficiency of a cold-side ESP. However,  $\text{SO}_3$  usually increases the deposits and corrosion in air preheaters (APH) and gas-gas heaters (GGH).

The oxidation can be maintained at a low level (below 0.5%) by using a low  $\text{SO}_2$  oxidation catalyst and operating at a relatively low temperature (below about 350°C). In such a situation, the catalyst may not be very active and a considerable amount of unreacted  $\text{NH}_3$  (slip  $\text{NH}_3$ ) may be present at the reactor outlet resulting in the deposits of ammonium bisulfate in the APH.

On the other hand, by using an active catalyst at a higher temperature, slip  $\text{NH}_3$  can be reduced below 3 ppm but  $\text{SO}_3$  may exceed 10 ppm when the gas is rich in  $\text{SO}_2$ . Most of the  $\text{SO}_3$  is absorbed by fly ash but  $\text{SO}_3$  in the gas phase at the GGH inlet exceeding 3 ppm, may result in the deposit of sulfuric acid in the GGH, which causes corrosion and plugging. To solve the above problems, a larger volume of less active low oxidation catalyst should be used at 350°C or below.

With the low-dust system (Figure 9B), the catalyst erosion is less but a larger amount of  $\text{SO}_3$  may be present in the gas phase and thus deposits may form in the APH and GGH. The hot ESP is efficient for fly ash from low-sulfur coals, but costly. The low-dust system has not become popular for these reasons.

### 3.2 FGD FOLLOWED BY SCR (TAIL GAS TREATMENT)

Systems with FGD followed by SCR (Figure 9C-G) are also in commercial use to remove  $\text{SO}_2$  and  $\text{NO}_x$ . Systems C and D were put into commercial operation in Japan in 1975 and 1976, respectively, when  $\text{SO}_2$  resistant parallel-flow SCR catalysts were not yet well developed.

The system shown in Figure 9 C was used for an oil-fired boiler with a flue gas flow rate of 440,000  $\text{Nm}^3/\text{h}$ . It had corrosion and plugging problems with the GGH, because the gas leaving the wet FGD plant contained about 30 ppm  $\text{SO}_2$  and about 100 ppm  $\text{SO}_3$ . The system was shut down

after several years operation and was replaced by a new system, high-dust SCR followed by FGD, which has been performing well.

The system shown in Figure 9 D is for a 762,000 Nm<sup>3</sup>/h flue gas at 150°C from an iron-ore sintering machine at the Chiba Works of Kawasaki Steel. The system has operated well because it has a wet ESP downstream of the wet FGD to remove particulate matter and SO<sub>2</sub>. A similar system has also been applied to 250°C flue gases from 15 sewage sludge incinerators. It has been working well while also reducing the offensive odor caused by the SCR system. These systems require a lot of energy for the gas heating. The direct application of SCR to the flue gases at 150-250°C causes ammonium bisulfate deposit on the SCR catalyst.

The systems shown in Figure 9 E, F, and G, are used in FRG. System E is under construction at the Mannheim plant of Grosskraftwerk (GKM) at a 220 MW wet bottom coal-fired boiler. It is similar to system C and may have troubles with the GGH if the inlet gas contains a considerable amount of sulfuric acid mist.

The system shown in Figure 9 F has been applied since late 1987 at the Nuremberg plant of Energy and Wasserversorgung AG (EWAG) at three coal-fired boilers each producing 155,000 Nm<sup>3</sup>/h flue gas. It is a combination of lime slurry spray drying FGD, a special type heat regenerator, and SCR. The system is relatively economical because only a 20°C temperature rise is required to be produced by fuel.

The system shown in Figure 9 G has been used at the Neuburg plant of Oberland Glass since 1987 to treat 50,000 Nm<sup>3</sup>/h of flue gas from a glass melting furnace. The gas at 450°C is sprayed with a slurry of water and slaked lime and then enters a hot ESP at 400°C and is then subjected to SCR. The system removes about 55% of the SO<sub>2</sub>, more than 90% of the SO<sub>3</sub>, 60-80% of the NO<sub>x</sub>, and 99% of the dust and seems to have been working well.

### 3.3 SIMULTANEOUS SO<sub>2</sub> AND NO<sub>x</sub> REMOVAL

#### Activated Coke Process

Two simultaneous SO<sub>2</sub>/NO<sub>x</sub> removal plants using activated coke in a moving bed have been operated since 1987. One was constructed by Mitsui Mining for Idemitsu Kosan at the Aichi oil refinery to treat 236,000 Nm<sup>3</sup>/h of flue gas from a Fluid Catalytic Cracking (FCC) plant containing 340 ppm SO<sub>2</sub>, 150 ppm NO<sub>x</sub>, and 60 mg/Nm<sup>3</sup> of dust. It uses a two-stage reactor with addition of ammonia to the second stage, and removes 99% of the SO<sub>2</sub>, 70% of the NO<sub>x</sub>, and 50% of the dust that would otherwise have been emitted. The activated coke containing adsorbed SO<sub>2</sub> is regenerated by indirect heat, producing concentrated SO<sub>2</sub> gas which is introduced into an existing Claus furnace to produce elemental sulfur. The other plant was constructed by Sumitomo Heavy Industries for Nippon Steel at the Nagoya Works to treat 900,000 Nm<sup>3</sup>/h of flue gas from an iron-ore sintering machine containing 140 ppm SO<sub>2</sub> and 230 ppm NO<sub>x</sub>. By using a single reactor and a small amount of ammonia, about 30% of the NO<sub>x</sub> and 99% of the SO<sub>2</sub> are removed to meet the local regulations. The recovered SO<sub>2</sub> is introduced into an existing sulfuric acid plant. These two plants have operated well.

EPDC has also conducted large scale tests on an activated coke process to by-produce elemental sulfur (6). There is also an activated coke process plant in FRG. These plants had some initial problems which seem to have been solved. The activated coke processes are probably not well suited to gases containing high concentrations of SO<sub>2</sub>, since coke consumption increases with the SO<sub>2</sub> content of the gas.

#### Other Dry Processes

The Electron Beam (E-Beam) process originally developed by Ebara Corporation has been further developed in the U.S. and FRG using pilot plants with capacities of 10,000-20,000 Nm<sup>3</sup>/h

of flue gas from coal-fired boilers. About 90% of the  $\text{SO}_2$  and 80% of the  $\text{NO}_x$  are removed, by-producing an ammonium sulfate/nitrate mixture which is useful as fertilizer (7). The plant cost is estimated to be about equal to that of the combined SCR-FGD system but the power consumption reaches 3%. The process treats a flue gas below  $150^\circ\text{C}$  and may be suitable for retrofit to existing boilers and furnaces provided that a larger E-Beam accelerator becomes available at a reasonable price and the power consumption is reduced below 2%. Large scale demonstration plants for coal-fired boilers are expected to be constructed in FRG.

The Asahi Glass Company has used a unique process to treat  $87,000 \text{ Nm}^3/\text{h}$  of flue gas at  $400\text{--}450^\circ\text{C}$  from a glass melting furnace containing 1500 ppm  $\text{SO}_2$ , 400 ppm  $\text{NO}_x$ , and 300 mg/ $\text{Nm}^3$  of dust. About 0.35 mol  $\text{NH}_3$  to 1 mol  $\text{NO}_x$  is injected into the gas and then a sodium hydroxide solution is sprayed into the gas to by-produce dry sodium sulfate which is removed in an ESP. About 30% of the  $\text{NO}_x$  and 80% of  $\text{SO}_2$  are removed simultaneously. The by-product contains no nitrogen compounds, indicating that the  $\text{NH}_3$  probably reacts with  $\text{NO}_x$  to produce  $\text{N}_2$ , similar to the SCR reactions. The by-product sulfate is refined and fed to the glass furnace.

#### Wet Processes

About 10 commercial wet simultaneous  $\text{SO}_2/\text{NO}_x$  removal plants have been operated with unit capacities of  $10,000\text{--}200,000 \text{ Nm}^3/\text{h}$ . These processes employ an oxidizing agent,  $\text{ClO}_2$  or  $\text{O}_3$ , which is added to the gas, before it is subjected to wet scrubbing to remove 30–80% of the  $\text{NO}_x$  and more than 90% of the  $\text{SO}_2$ . These processes are not suitable to treat large volumes of gas because of the high cost of the oxidizing agent and the problems of disposing of wastewater containing nitrite and nitrate.

### 4. GAS CLEANING FOR MUNICIPAL SOLID WASTE INCINERATORS IN JAPAN

#### 4.1 OVERVIEW

Municipal solid waste (MSW) is generated in Japan at about 120,000 metric tons/day, with about 70% burned and the remainder, primarily disposed of in landfills. MSW produces about  $6,000\text{--}14,000 \text{ Nm}^3$  of combustion gas containing 40–100 ppm  $\text{SO}_2$ , 150–250 ppm  $\text{NO}_x$ , 300–800 ppm  $\text{HCl}$ , 8–14%  $\text{O}_2$ , and 1–3 g/ $\text{Nm}^3$  of dust per metric ton of waste. The MSW incinerators in Japan with unit capacities larger than 1 metric ton/h may number about 2000. The largest one has a unit capacity of 17 metric tons/h or 400 metric tons/day. Each incineration plant usually has two or three incinerators.

A survey was conducted on 71 plants with 161 incinerators having unit capacities of 1.5–17 metric tons/h and made by 13 major manufacturers. Stoker furnaces are used at 50 plants, fluidized-bed furnaces at 19 plants, and rotary kilns at 2 plants. Electric power is generated at 31 plants. The power generation capacity of these plants ranges from 500 to 15,000 kW. The incinerators have ESPs for dust removal except for two plants with four incinerators that use fabric filters. The fly ash as well as the bottom ash is usually landfilled. Although the ashes contain hazardous components such as chlorinated organic compounds, mercury, and cadmium, these components are present at very low concentrations or are nearly insoluble in water. For an added measure of prevention against leaching, the fly ash is pelletized using cement at 20 plants. Most plants remove the acid gas ( $\text{SO}_2$  and  $\text{HCl}$ ) by spray drying a lime slurry or by dry slaked lime injection in the gas followed by dry collection. Wet sodium scrubbing is applied at 21 medium size plants. Table 3 shows the investment cost of the plants including civil works, pollution control facilities, and office buildings. The cost ranges from 200 to 700 million yen/metric ton/h ( $\$1600\text{--}\$5600/\text{metric ton/h}$ ) according to the size and type of plant.

#### 4.2 GAS CLEANING SYSTEMS FOR MUNICIPAL WASTE INCINERATORS

$\text{NO}_x$  emissions are usually reduced from 150–250 to 70–150 ppm (at 12%  $\text{O}_2$  basis) by combustion modification including the reduction of  $\text{O}_2$  content of the gas, two-stage combustion,



and the lowering of the combustion temperature from 900-1000 to 700-900°C. Water is usually injected into the furnace to lower the temperature.

At some of the fluidized-bed furnaces, limestone or dolomite is used as a bed material. Pulverized limestone is injected into the furnace at around 900°C at a few large plants (Figure 10, A). By those methods, about 50% of both  $\text{SO}_2$  and HCl are removed at a lime stoichiometry,  $\text{CaO}/(\text{SO}_2 + 2\text{HCl})$  mole ratio, of about 2.

At 33 plants with 78 incinerators, dry slaked lime or slurry is injected or sprayed into a duct or a reactor at 250-400°C to remove 70-98% of  $\text{SO}_2$  and HCl (Figure 10, B, D, and H) using stoichiometries of 2-2.5. The efficiencies are higher with slaked lime than with limestone, with slaked lime slurry than with dry slaked lime, and with a fabric filter than with an ESP.

Wet sodium scrubbing is used at 21 plants with 44 incinerators to remove 70-98% of  $\text{SO}_2$  and HCl. At most plants, ESPs are placed upstream of the wet scrubber (Figure 10, C). At one plant, a wet ESP is placed downstream of the wet scrubber to reduce dust below 3 mg/Nm<sup>3</sup> and  $\text{SO}_2$  and HCl below 3 ppm.

For  $\text{NO}_x$  removal, SNR is applied at four plants (Figure 10 D and E) to remove 30-40% of  $\text{NO}_x$  using 1-1.5 mole  $\text{NH}_3$  per 1 mole  $\text{NO}_x$  (Figure 7)--a larger amount of  $\text{NH}_3$  tends to cause a plume problem. The efficiency is lower than with an oil-fired boiler, because the fluctuations are larger in both gas temperature and  $\text{NO}_x$  concentration. At two plants, urea solution is sprayed into the furnace to reduce emissions by about 30%  $\text{NO}_x$  (Figure 10, F and I). SCR is used at two plants to reduce  $\text{NO}_x$  emissions by 50-80% at 200-250°C using an HCl-resistant catalyst (Figure 10, G and H).

#### 4.3 MERCURY AND DIOXINS

Mercury is derived mainly from electric cells and is present in concentrations of 0.03-0.3 mg/Nm<sup>3</sup> in MSW incineration gases. Wet scrubbing removes 60-80% of the mercury while dry processes remove less. Thus, waste gases from MSW incineration plants contain a small amount of mercury mainly in the form of metallic vapor. Methyl mercury, the most harmful compound, is not detected in the gases.

At two plants more than 95% of the mercury is removed by sodium scrubbing using  $\text{NaClO}$  as the additive. A portion of the  $\text{NO}_x$  is also removed by the scrubbing. Mercury in the scrubber liquor is usually precipitated and removed from waste water. Recovery of metallic mercury from the scrubber liquor has started.

Dioxins are usually contained in the incineration gas at concentrations of 0.1-1  $\mu\text{g}/\text{Nm}^3$ . The concentration is less when a large amount of excess air is used for the incineration. Most of the dioxins are removed by conventional flue gas cleaning systems. The waste gas from MSW incineration plants usually contains 0.01-0.5  $\mu\text{g}/\text{Nm}^3$  of dioxins but the most harmful 2,3,7,8 tetrachloro-dibenzoparadioxin concentration is less than 0.01  $\mu\text{g}/\text{Nm}^3$ , a level which may not cause environmental problems.

### 5. $\text{SO}_2$ AND $\text{NO}_x$ REMOVAL IN CHINA

The first commercial FGD plant in China may be the 52,000 Nm<sup>3</sup>/h unit constructed in 1978 near Nanjing for a flue gas from a pelletizing machine. The plant reduces  $\text{SO}_2$  concentration from 2800 to 100 ppm using the Dowa aluminum sulfate limestone process to by-produce gypsum.

Three ammonia scrubbing plants were constructed in 1987 by IHI for three oil-fired boilers at different locations including Nanjing and Shanghai. Each plant has a capacity for treating 220,000 Nm<sup>3</sup>/h flue gas to reduce  $\text{SO}_2$  concentration from 1200 to 120 ppm or below. Two absorbers in series are used to minimize the plume problem common to ammonia scrubbing. It is



likely that a few other FGD plants have been constructed recently, including one Niro atomizer spray drying process.

Huaneng International Power Development Co. has decided to install limestone-gypsum process plants for two 360 MW coal-fired boilers under construction in Chongqing to reduce  $\text{SO}_2$  emissions from 4700 to 230 ppm. MHI has started construction of the plants using the Figure 3, type IX scheme. These FGD plants will be put into operation in 1990 and 1991, respectively.

The Chinese generally prefer FGD processes yielding useful by-products, because China has a large population and limited land area to dispose of waste products over a long period. Sulfuric acid and ammonium sulfate are desirable by-products. Gypsum is useful for cement manufacture, but there is no current market for wallboard.

Small scale tests have been made on activated coke processes which by-produce sulfuric acid by water-wash of the coke loaded with  $\text{SO}_2$ . The Nuclear Research Institute in Shanghai has started research on the E-Beam process that by-produces ammonium sulfate/nitrate fertilizer.

China has many small fluidized bed combustion boilers and furnaces using coal but these have not been designed for  $\text{SO}_2$  abatement. The addition of limestone or dolomite as the bed material as well as pulverized limestone injection into the furnace may be useful provided that the by-product can be used. A possible use may be for road paving--most roads in China are unpaved. A model fluidized bed combustion plant using coal and limestone as the bed material has started operation in Nanning.

SCR has been applied to a nitric acid plant tail gas since 1980 using a pellet catalyst developed in China. Two SCR units have been operated with a unit capacity of treating 90,000  $\text{Nm}^3/\text{h}$  of the tail gas to reduce  $\text{NO}_x$  from 3000-4000 to 500 ppm. Although the SCR units have worked well, a large amount of ammonia is consumed. It may be preferable to modify the acid plants to reduce  $\text{NO}_x$  in the tail gas below 1000 ppm and then apply SCR if necessary.

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## 7. ABBREVIATIONS

### Company Names:

BHK: Babcock Hitachi K.K.  
EPDC: Electric Power Development Co.  
EWAG: Energy and Wasserversorgung AG  
GKM: Grosskraftwerk-Mannheim  
IHI: Ishikawakima-Harima Heavy Industries  
KHI: Kawasaki Heavy Industries  
MHI: Mitsubishi Heavy Industries  
MEC: Mitsui Miike Engineering Co.

### Others:

|                                         |                                      |
|-----------------------------------------|--------------------------------------|
| APH: Air preheater                      | CM: Combustion modification          |
| ESP: Electrostatic precipitator         | GGH: Gas-gas heater                  |
| IFR: In-furnace $\text{NO}_x$ reduction | MSW: Municipal solid waste           |
| MW: Megawatt (electrical)               | Nm <sup>3</sup> : Normal cubic meter |
| SCR: Selective catalytic reduction      | $\mu\text{m}$ : micrometer           |
| SNR: Selective noncatalytic reduction   | $\mu\text{g}$ : microgram            |

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.

TABLE 1. COAL-FIRED UTILITY BOILERS AND FLUE GAS CLEANING SYSTEMS  
(In Japan, by 1987)

| Type  | FGD | SCR | Number | Capacity<br>(MW) | Capacity<br>(percent of total) |
|-------|-----|-----|--------|------------------|--------------------------------|
| A     | No  | No  | 11     | 1,200            | 10                             |
| B     | Yes | No  | 11     | 3,286            | 28                             |
| C     | Yes | Yes | 26     | 7,354            | 62                             |
| Total |     |     | 48     | 11,840           | 100                            |

TABLE 2. FGD AND SCR UNITS FOR COAL-FIRED UTILITY BOILERS (1983-1993)

| Owner             | Station<br>Name | Boiler<br>(MW) | FGD         |            |      | SCR         |            |      |
|-------------------|-----------------|----------------|-------------|------------|------|-------------|------------|------|
|                   |                 |                | Constructor | Completion | Type | Constructor | Completion | Type |
| EPDC              | Takehara        | 700            | IHI         | 1983       | III  | BEK         | 1983       | L*   |
| Johan Joint       | Nakoso          | 600 x 2        | MHI         | 1983       | IV   | MHI, IHI    | 1983       | F    |
| Kyushu Electric   | Minato          | 156            | MMBC        | 1983       | VII  | MHI         | 1983       | H    |
| Shinkoku Electric | Saijo           | 156            | KHI         | 1983       | VI   | MHI         | 1983       | H    |
| Toboku Electric   | Sendai          | 175 x 2        | BEK         | 1983       | IV   | BEK         | 1983       | H    |
| Shikoku Electric  | Saijo           | 250            | KHI         | 1984       | VI   | IHI         | 1984       | H    |
| Chugoku Electric  | Mizushima       | 125 & 156      | BEK         | 1984       | IV   | BEK         | 1984       | H    |
| Sakata Joint      | Sakata          | 350            | MHI         | 1984       | IV   | MHI         | 1984       | H    |
| Toyama Joint      | Toyama          | 200 x 2        | Chiyoda     | 1984       | V    | BEK         | 1984       | H    |
| Tokyo Electric    | Yokosuka        | 265*           | MHI         | 1984       | IV   | MHI         | 1984       | H    |
| Tokyo Electric    | Yokosuka        | 265*           | BEK         | 1985       | IV   | MHI         | 1985       | H    |
| Hokkaido Electric | Tomato          | 600            | MHI         | 1985       | IV   | None        | -          | -    |
| Chugoku Electric  | Shin-Onoda      | 500            | IHI         | 1986       | IV   | IHI         | 1986       | H    |
| EPDC              | Ishikawa        | 125            | IHI, MMBC   | 1986       | III  | None        | -          | -    |
| Chugoku Electric  | Shin-Onoda      | 500            | IHI         | 1987       | IV   | IHI         | 1987       | -    |
| EPDC              | Ishikawa        | 125            | IHI, MMBC   | 1987       | VII  | None        | -          | -    |
| Kyushu Electric   | Matsuyama       | 700            | MHI         | 1989       | VIII | MHI         | 1989       | H    |
| EPDC              | Matsuyama       | 1000           | BEK         | 1990       | IX   | BEK         | 1990       | L    |
| Hokuriku Electric | Tsuruga         | 500            | NU*         | 1991       | ND   | ND          | 1991       | ND   |
| Chubu Electric    | Bekinan         | 700            | MHI         | 1991       | VIII | MHI         | 1991       | H    |
| Chubu Electric    | Bekinan         | 700            | KHI         | 1992       | IX   | BEK         | 1992       | H    |
| Chubu Electric    | Bekinan         | 700            | Chiyoda     | 1993       | V    | IHI         | 1993       | H    |

\* Low-dust system

\* Burning coal-oil mixture

\* Burning coal and oil

\* Not determined

\* High-dust system

TABLE 3. INVESTMENT COST OF INCINERATION PLANTS  
(10<sup>4</sup> yen/metric ton/h, \$1 = 125 yen)

| Scrubbing type | Large plants* | Medium plants* | Small plants** |
|----------------|---------------|----------------|----------------|
| Dry, Semi-dry  | 200 - 300     | 300 - 600      | 200 - 500      |
| Wet (sodium)   |               | 400 - 700      | 400 - 700      |

\*With power generation

\*\*Without power generation

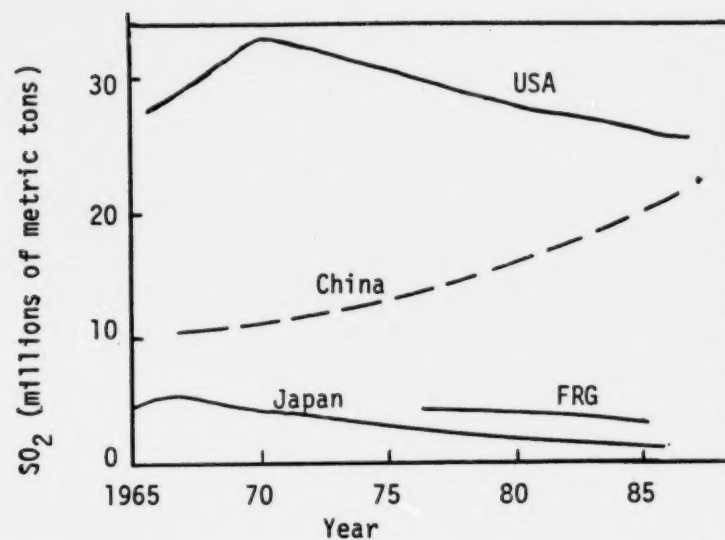


Figure 1. Yearly SO<sub>2</sub> emissions



Figure 2. Rain pH in China (average May-August 1982)



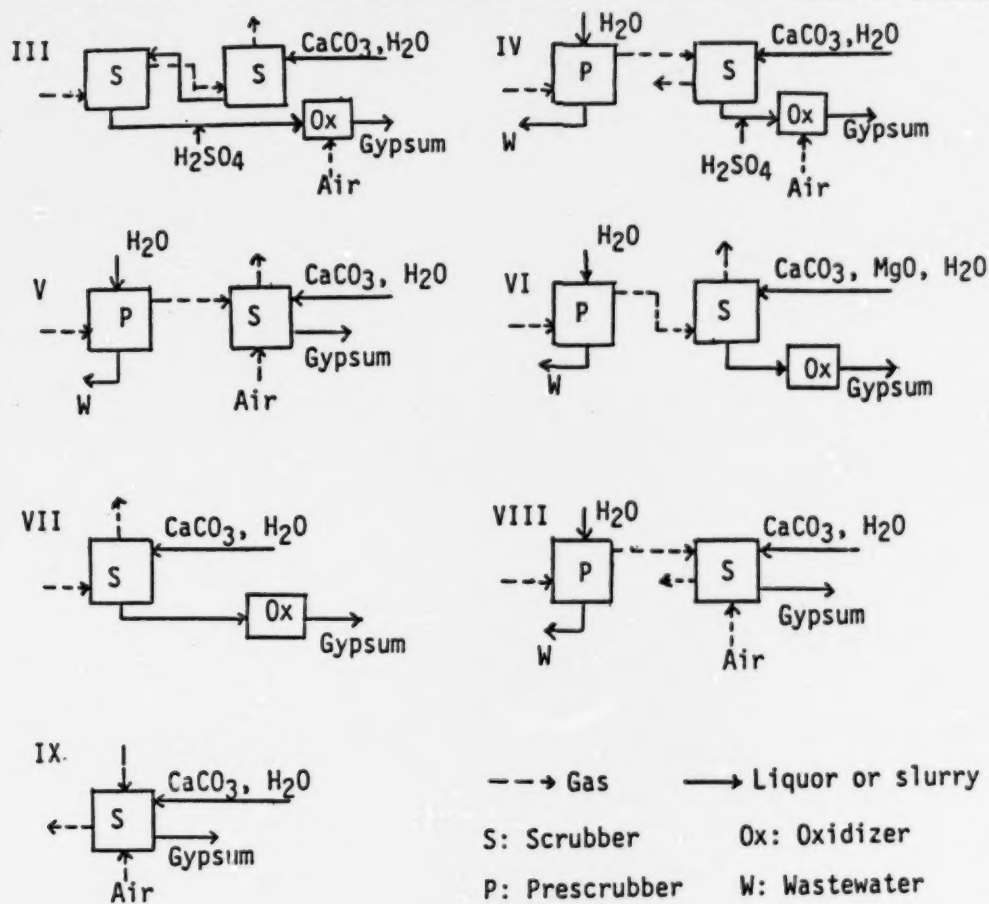


Figure 3. Wet limestone-gypsum processes

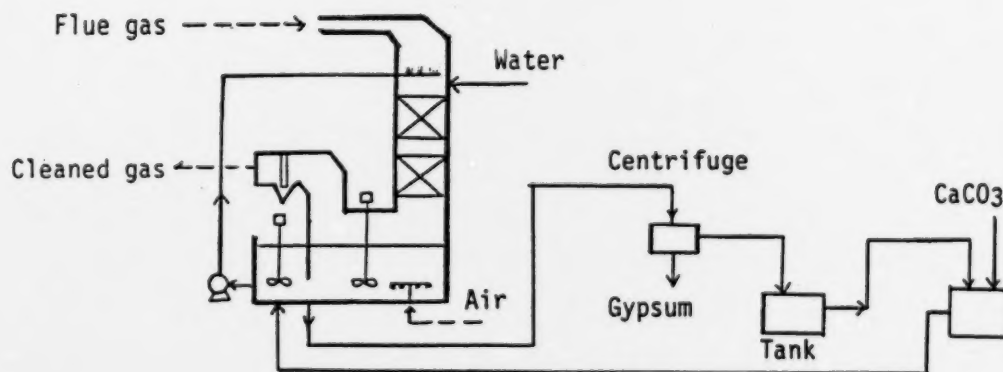


Figure 4. Type IX plant by MHI

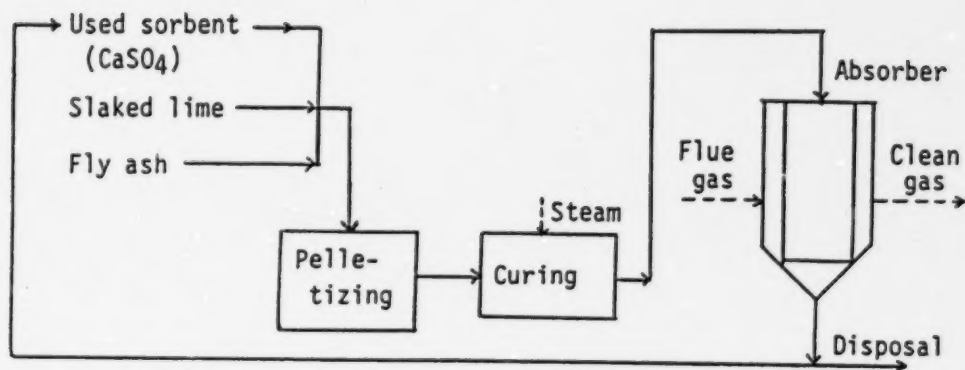


Figure 5. Dry lime fly ash based sorbent process by Hokkaido Electric

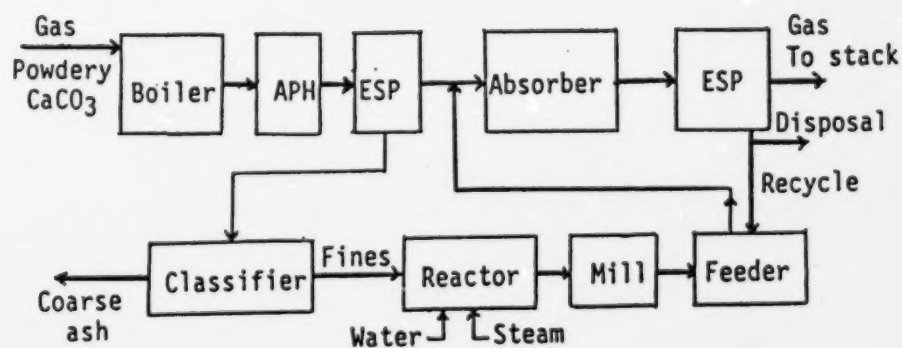


Figure 6. Dry powdery sorbent injection process

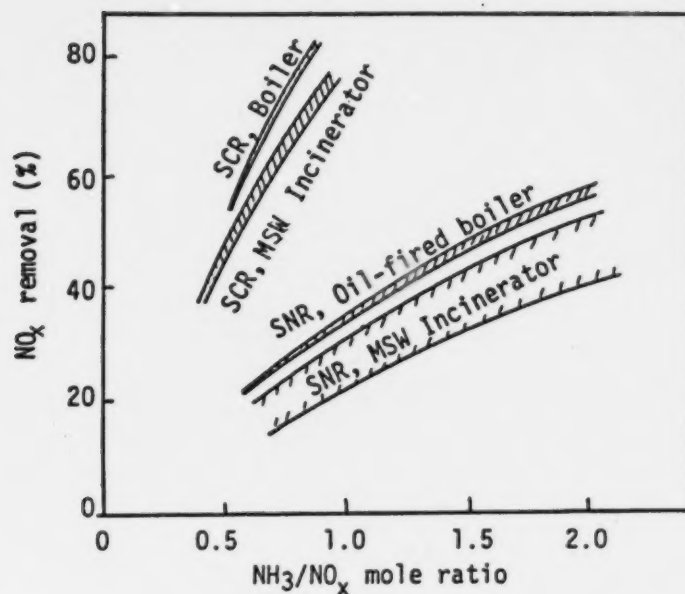
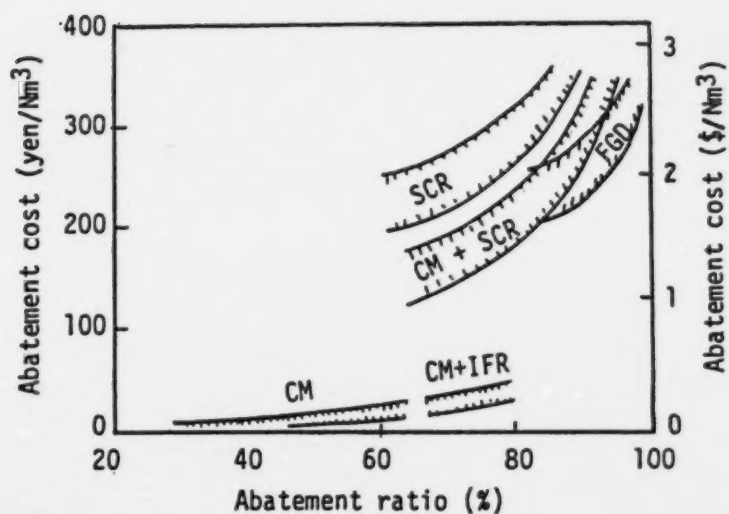


Figure 7.  $\text{NO}_x$  removal by SCR and SNR

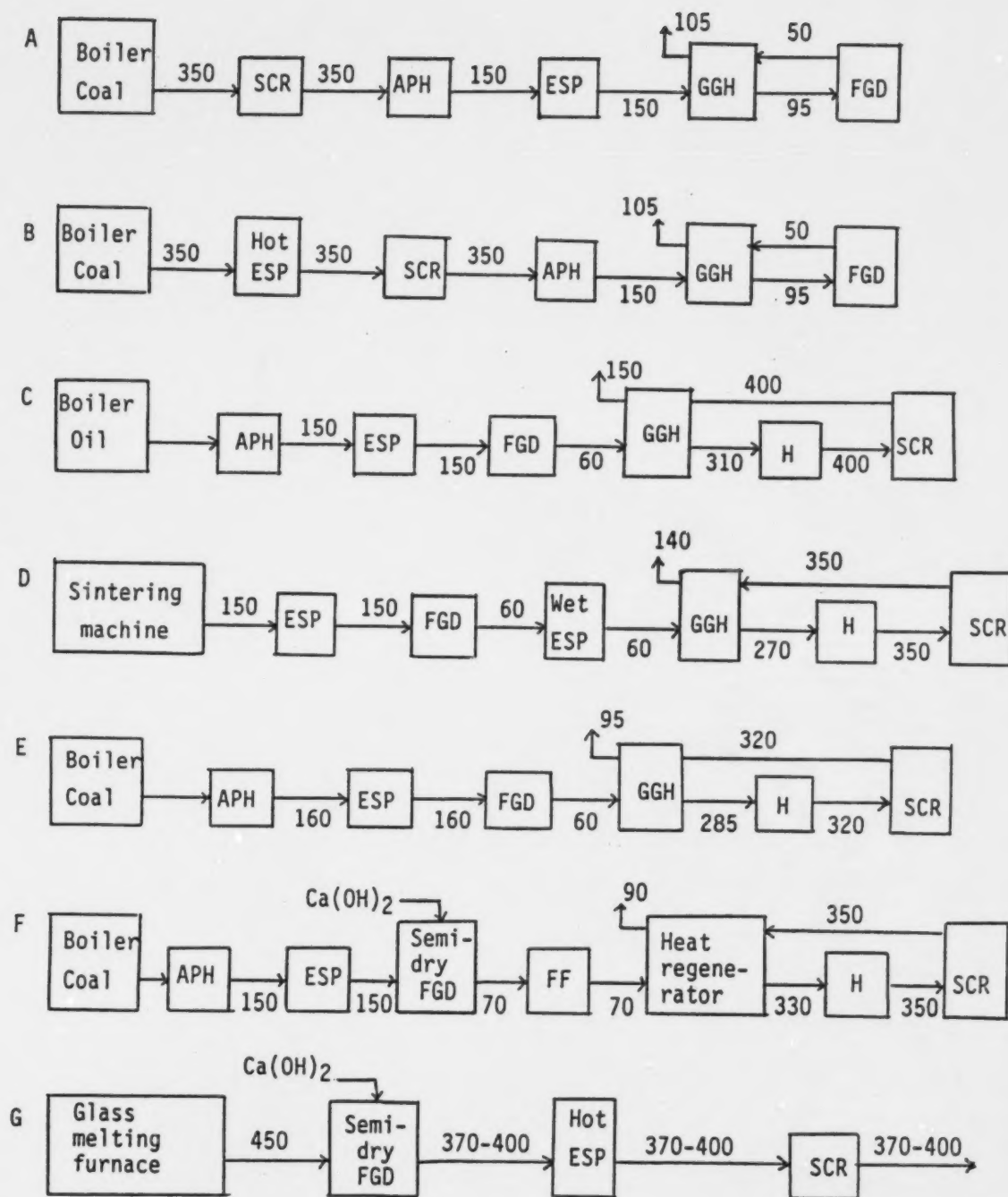


FGD: Wet limestone-gypsum process  
 CM: Combustion modification  
 IFR: In-furnace  $\text{NO}_x$  reduction  
 Coal-fired utility boiler  
 $\text{SO}_2$  1000 ppm,  $\text{NO}_x$  600 ppm (uncontrolled)

$1 \text{ Nm}^3 \text{ SO}_2 = 2.86 \text{ kg}$

$1 \text{ Nm}^3 \text{ NO}_x = 2.05 \text{ kg}$

Figure 8. Estimated  $\text{SO}_2$  and  $\text{NO}_x$  abatement cost



APH: Air preheater    ESP: Electrostatic precipitator  
GGH: Gas-gas heater    H: Heater    FF: Fabric filter

Figure 9. SCR-FGD and FGD-SCR combined systems in Japan (A-D) and FRG (A,E,F,G)



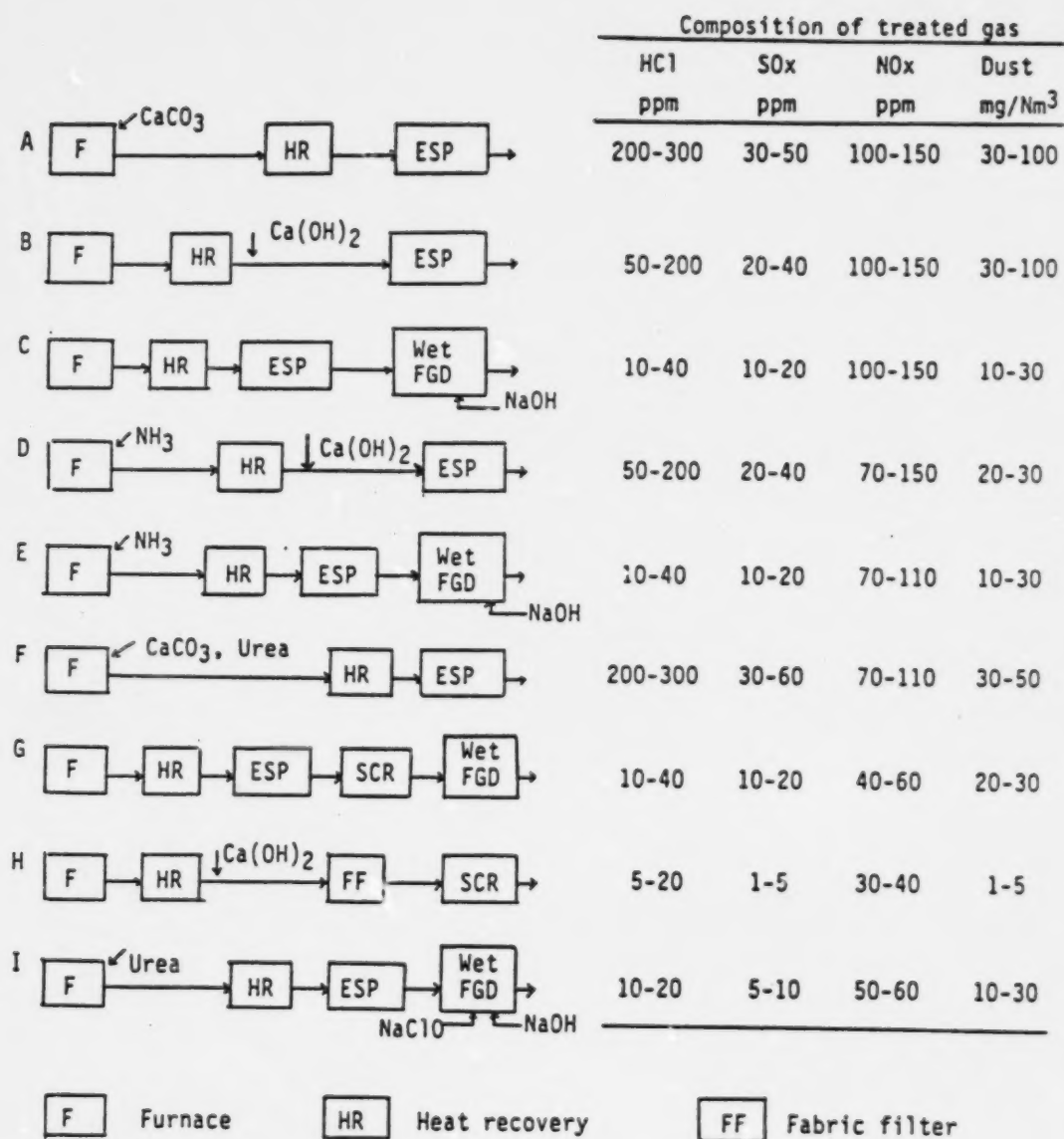


Figure 10. Flue gas cleaning systems for MSW incinerators

## SESSION 2: RETROFIT ECONOMICS

Chairman, Paul T. Radcliffe, EPRI

### THE DESIGN, OPERATION, AND PERFORMANCE OF CURRENT GENERATION RETROFIT FGD SYSTEMS ON COAL-FIRED UTILITY BOILERS

B. A. Laseke  
PEI Associates, Inc.  
11499 Chester Road  
Cincinnati, Ohio 45246

First Combined FGD and Dry SO<sub>2</sub> Control Symposium

Sponsored by:  
U. S. Environmental Protection Agency  
Air and Energy Engineering Research Laboratory  
and  
Electric Power Research Institute  
Coal Combustion Systems Division

October 25-28, 1988  
Adam's Mark Hotel  
St. Louis, Missouri

#### ABSTRACT

Flue gas desulfurization (FGD) represents the most commercially-mature technology in the technology slate of SO<sub>2</sub> controls for coal-fired utility boilers. Accordingly, FGD (wet and dry) represents the technology of primary consideration and the benchmark technology of comparison for retrofit controls in anticipation of acid rain emission reductions. Site-specific cost and performance estimates have been prepared for existing plants in the top SO<sub>2</sub>-emitting population. These estimates have been prepared for projects of national scope (National Acid Precipitation Assessment Program) and regional scope (State Acid Rain). These estimates were prepared with the aid of existing computerized cost models and retrofit guidelines. One method of benchmarking these estimates is to compare them to the costs of retrofit FGD systems that are representative of current generation technology in terms of design, performance, and operation. A number of such systems meeting these criteria are in service. These FGD systems are analyzed with respect to retrofit design philosophy, design and performance characteristics, and costs (total retrofit capital and annual). A comparison of retrofit FGD to new plant FGD technology through an analysis of retrofit difficulty is presented. An assessment of the ability to predict FGD costs using existing models and retrofit procedures is presented.

## BACKGROUND

### INTRODUCTION

During the past five years, PEI Associates, Inc. (PEI) has evaluated the potential application of air emission control technologies to existing coal-fired utility boilers in the United States. The purpose of these investigations was to provide cost and performance estimates for the control of acidic emissions by retrofit emission control technologies. These investigations were conducted as separate research efforts funded primarily through the State Acid Rain (STAR) research program and the National Acid Precipitation Assessment Program (NAPAP). The common objective of these studies (Table 1) was to make significant improvement in current site-specific cost and performance estimates for a wide variety of coal-fired power plant emission control options. This information, in turn, was used by policy makers to evaluate and develop acid rain control strategies.

### PURPOSE

Flue gas desulfurization (FGD) represents the most commercially-mature technology in the technology slate of SO<sub>2</sub> controls for coal-fired utility boilers. Accordingly, FGD (wet and dry) represents the technology of primary consideration and the benchmark technology of comparison for retrofit controls in anticipation of acid rain emission reductions. Site-specific cost and performance estimates were prepared for existing plants in the top SO<sub>2</sub>-emitting population. One method of benchmarking these estimates is to compare them to the costs of retrofit FGD systems that are representative of current generation technology in terms of design, performance, and operation. A number of such systems meeting these criteria are in service. These FGD systems are analyzed herein with respect to retrofit design philosophy, design and performance characteristics and costs (total retrofit capital and annual). A comparison of retrofit FGD to new plant FGD technology through an analysis of retrofit difficulty is presented. An assessment of the ability to predict FGD costs using existing models and retrofit procedures is presented.

### TECHNICAL APPROACH

The technical approach adopted for these investigations involved the collection of site-specific design, operating, and performance data to support technology and process selection, cost and performance assumptions, and assignment of retrofit difficulty factors. These data needs were satisfied through the development of a computerized data base containing boiler-specific design, operating, fuel, performance, and emissions control data. This computerized data base was interfaced with a computerized cost model to generate site-specific cost and performance estimates. The cost model is a computer program that integrates air emission control technologies through cost and performance interactions. This model was originally developed by PEI and has been subsequently modified for retrofit applications.

### DESCRIPTION OF DATA BASE

The boiler data base developed by PEI to support retrofit emission control technology assessments originated from a larger data base of coal-fired utility boilers also developed by PEI known as Acidbase. Acidbase contains design, performance, operating, and emissions data for all coal-fired utility boilers in commercial service for the years of 1982 to 1985. The sources of existing information pertinent to coal-fired utility boilers that were immediately accessible for the development of Acidbase included public data bases maintained by the Federal Government. The more pertinent sources of information accessed were the DOE energy data bases and EPA environmental data bases. The EIA Form 767 was determined to be the most recent and comprehensive source of information for coal-fired utility boilers. Computer tapes of the 1982 through 1985 EIA Form 767 submissions were obtained. The data from the mainframe computer tapes were downloaded and ported into a PC using PC-based data base management system. The data were then quality checked for accuracy and completeness. Selected data elements for the record year of 1985 were extracted from Acidbase to create a subset data base known as Coalbase which was then interfaced with the cost model. These data elements were selected based on their

relevance to the evaluation process. Specific data elements include descriptive characteristics (ORIS, utility, plant, unit, boiler, state, region), boiler characteristics (supplier, rating, firing type, steam cycle, furnace dimensions), fuel characteristics (primary and secondary fuels, coal rank, coal heat content, coal approximate analysis), unit operating characteristics (status, duty cycle, service hours, capacity factor), and emission controls (control systems, SO<sub>2</sub> and NO<sub>x</sub> emissions).

The sources of existing information pertinent to coal-fired utility boilers that were immediately accessible for the development of Acidbase included public data bases maintained by the Federal Government. The more pertinent sources of information accessed were the DOE energy data bases and EPA environmental data bases. The EIA Form 767 was determined to be the most recent and comprehensive source of information for coal-fired utility boilers. Computer tapes of the 1982 through 1985 EIA Form 767 submissions were obtained. The data from the mainframe computer tapes were downloaded and ported into a PC using PC-based data base management system. The data were then quality checked for accuracy and completeness.

A listing of the data elements contained in Coalbase is shown in Table 2.

## DESCRIPTION OF MODEL

### BACKGROUND

The Integrated Air Pollution Control System (IAPCS) was developed to estimate the costs and predict the performance of sulfur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>), and particulate matter (PM) emission control systems for coal-fired utility boilers. The model includes conventional and emerging technologies that effect pre-, in situ, and post-combustion emission control. The model can accept any combination of the technology "modules" built into the system. Interactions are reflected in a material balance tabulation at the exit of each module. Alterations in the material balance are used to account for integrated performance and cost effects. The emission control technologies contained in IAPCS can be selected in either "isolated" (single technology) or "integrated" (multiple technology) configurations. The power of IAPCS lies in its ability to reflect integrated effects of various control configurations. This allows the analyst to identify synergistic interactions and thus optimize performance and cost in terms of integrated cost effectiveness.

The first version of IAPCS (IAPCS-I) was completed in November 1983<sup>(1)</sup>. This version was a mainframe computer model housed at EPA's National Computer Center (NCC). The second version of IAPCS (IAPCS-II) was completed in July 1986<sup>(2)</sup>. This version incorporated a number of enhancements to the design premises of the pollution control modules as well as the model's user access and versatility. A third version of IAPCS (IAPCS-III) has been recently completed by PEI. This version was prepared to support ongoing work in acid rain control technology assessment. The overall rationale for the development of IAPCS-III was the need to adapt the model to the retrofit conditions associated with existing coal-fired utility boilers as well as the desire to improve user friendliness and significantly increase processing speed. This version incorporates a number of enhancements in the form of additional technology modules, improvements to and expansion of existing technology modules, and program modifications to increase processing speed for batch processing.

### OVERVIEW

The IAPCS performance and cost-estimating computer model was developed to estimate the cost and performance of air emission control equipment for coal-fired utility boilers. The model includes both conventional and emerging control technologies (Table 3).

As designed, the model accepts any combination of these technologies. System interactions are reflected in a material balance tabulation at the exit of each module. An "uncontrolled" material balance column is calculated before the pre- and in situ control modules are applied so that the net effect of emission control can be calculated on a system basis. Output from the model reports the reduction in SO<sub>2</sub>, PM, and NO<sub>x</sub>.



to SO<sub>2</sub> through a parameter file option. For PM, an 80/20 split of ash is assumed as the topside and bottom ash fractions in the calculations. In this case, the user has the option of applying the AP-42 emission factors contained in the parameter file. For NO<sub>x</sub> emissions, the AP-42 emission factors are used.

Boiler Performance--The net heat rate of the boiler is calculated to show the energy penalty imposed by the control system on the unit. The unit's thermal efficiency is based on the heating value of the coal. This thermal efficiency is used to adjust a minimum heat rate upward, and the losses to the system for auxiliaries are added (in Btu/kWh). The gross heat rate is then calculated, and power losses due to each of the control technology modules selected in the control configuration are added to the heat rate. In this case, the net heat rate reflects the total Btu/kWh required for the boiler and the selected control configuration.

Fans--Another system-level calculation routine (FANS) is provided to add booster fans for the selected integrated control configuration. The design of the induced-draft booster fans and the estimated costs are based on the total gas-side pressure drop and the gas flow rate. Many different control configurations are possible; therefore, the FANS module may be used to calculate forced-draft fan costs where appropriate.

Waste Disposal--The model provides three waste disposal options: all waste can be disposed of offsite; the waste can be split (in any ratio) between offsite and onsite; or all waste can be disposed of onsite. No unlined pond untreated waste options are provided.

Economics--The economics module provides economic, financial, and unit cost data for the computation of full scope capital costs, the computation of the O&M component of the annual cost, the computation of the carrying charge component of the annual cost, and the levelization factors used in present worth accounting.

Parameter File--The parameter file is a critical facet of the model. Through it, changes affecting input (run) conditions and the design, performance, and cost of individual modules may be facilitated. Access to this file permits the user to obtain maximum flexibility in depicting a given control scenario and to update and revise control technologies as data become available.

### Output Format and Options

The model provides the user with eight separate reports. They are highlighted below.

- User Input Summary--The first output is a reiteration of the inputs provided by the user.
- Module-Specific Output--Brief statements describing the primary design characteristics of the selected control modules.
- Boiler Performance--Heat input, boiler efficiency, net heat rate, and coal consumption are calculated by using the coal characteristics (ROM or cleaned).
- Material Balance--Material balance components are calculated at the exit of each control module. The "uncontrolled" column calculates a baseline estimate using the ROM coal characteristics so that the overall system pollutant reduction effects can be calculated; i.e., the uncontrolled column does not reflect any of the boiler-related (pre-combustion and in situ) controls.
- Emission Reduction--The overall system emission reduction is reported in a summary table. This table presents the mass flow rate (lb/h), percent removal, and unitized mass and volume emission rates (lb/10<sup>6</sup> Btu and ppm) for PM, SO<sub>2</sub>, and NO<sub>x</sub>.

- Cost Estimate--The capital and annual cost estimate for the designated control configuration is the next output. All capital indirects and annual O&M and capital cost components are identified and displayed.
- Cost Effectiveness--An output of the system's cost effectiveness (CE) is then provided. The cost per ton (\$/ton) of PM, SO<sub>2</sub>, and NO<sub>x</sub> removed is calculated for comparative purposes by using the levelized annual revenue requirement. The user should be cautioned that the cost effectiveness value displayed is calculated at the system level. Hence, it represents total dollars divided by total emissions removed (speciated by SO<sub>2</sub>, NO<sub>x</sub>, and PM). As such, these values are not "weighted" to specific emission reductions provided by specific technologies.

### Optimization

At the initiation of a run, the user can "optimize" a selected control system. A target emission rate (in lb/10<sup>6</sup> Btu) may be entered and the system performance and costs will be run automatically. This optimization routine allows the user to alter the effective efficiency of a chosen control device. If the user elects to optimize, the user will be prompted to enter a "target" emission rate, in pounds per million Btu, for the pollutant appropriate to the control module selected. (It is only a "target" because if other modules are in the system, they may also affect the final emission rate, and only one module is optimizable at a time.) The effective capture or removal efficiency of a technology module is changed either through bypassing a fraction of the gas stream or by simply changing the capture efficiency of the technology by altering its design and operating parameters.

### DESCRIPTION OF TECHNOLOGY MODULES

The IAPCS model is designed on a modular basis; i.e., a given control technology accepts the flue gas, coal, and unit characteristics from the previous module. These data are then used to generate the design, performance calculations, and estimates of capital and annual costs. The architecture of a modular program is such that it offers the user the greatest flexibility for revising any existing control device and for adding new technologies as they are identified.

The design and cost bases for each technology module are highlighted in Tables 4 through 14.

### INTEGRATED CHARACTERISTICS

The IAPCS model has been developed in part to provide a unique view of the performance of an air emission control system made up of individual modules. To this end, the performance of the entire system is output, as well as the material balance associated with each module in a specified control system. The material balance is the single most important integrated characteristic of the program. The relative significance of application of a given technology on a system basis can be readily assessed. Another integrated feature of the model involves the use of a system fan module. The individual pressure drops for any assembled control system are used to determine the overall horsepower and cost of the induced-draft fan(s). This is a less costly option for addressing the fan requirements than on a module-by-module basis. Another integrated feature is solid waste. Solid waste quantities are summed by module, and the cost of disposal of both wet and dry waste components are presented. Finally, the emission summary and the cost effectiveness outputs permit easy comparison of integrated control configurations from an economic standpoint.

### APPLICATIONS

The IAPCS model has been used extensively in a production mode for the past 3 years to generate site-specific cost and performance estimates for existing coal-fired utility boilers (Table 2). These projects have had one common theme--the application of retrofit controls to existing coal-fired utility boilers in anticipation of acid rain regulations limiting the emission of SO<sub>2</sub> and NO<sub>x</sub>. As part of these efforts, PEI prepared cost and performance estimates for a wide variety of retrofit controls to a large number of

emissions; associated capital and annual costs; and associated cost-effectiveness values (dollars per ton of pollutant removed across the entire emission control system).

A unique and important feature of the model is the parameter files. As each module was developed, the important design, cost, and performance parameters were included in a parameter file. These parameters are assigned default values that may be subsequently changed by the user for a given application. The parameter file is designed to permit the user to modify the important values to reflect those of choice or need.

## CAPABILITIES

A brief synopsis of IAPCS's capabilities, describing input features, processing, and output reports is presented below.

### Input Requirements

Inputs are provided either interactively through the parameter file or in a "batch" fashion through the data base. For PEI's projects, the batch file is provided by Coalbase. The batch file inputs either provide basic data for the given run or specifically affect the outcome of the run. Input requests include boiler data, fuel characteristics, and the control configuration. The boiler data are used to quantify the unit/system generating performance. The coal characteristics are used to estimate the emissions from firing a given quantity of coal, and the user specifies the controls to be utilized. The boiler firing type is used to estimate uncontrolled emissions.

### Cost Formats

The model incorporates the bases and format of cost estimation used by the Tennessee Valley Authority (TVA)<sup>(3)</sup> and the Electric Power Research Institute (EPRI)<sup>(4)</sup>, which are generally accepted as "industry standards". The capital cost is built "bottom-up", estimating direct capital costs which include direct and indirect construction costs. These costs are displayed for each process area comprised by the technology. Indirect capital cost components, which are added to the direct capital costs, are standardized and presented at the system level. Interpretation of the TVA and EPRI guidelines resulted in the assignment of percentages of the indirect components for each of the control modules.

As in the case of capital cost estimates, TVA and EPRI use different formats and bases to present annual costs. The maintenance labor and materials estimated by the TVA format for a given system represent percentages of the total direct investment. These same percentages (applied to process capital) are used to estimate maintenance labor and materials in the EPRI format. The costs of labor, reagents and chemicals, and waste disposal are specified in the TVA and EPRI cost formats. Calculations performed by the model yield the quantities of labor, additives and utilities, and waste generated to which the unit costs are applied to estimate the annual cost.

### System Files and Routines

Several of the files and routines are "system-wide" (i.e., not limited to one particular control technology module). These system-wide features are an important aspect of the model's integrated operation. They include standard coals, emission calculations, boiler performance, fans, waste disposal, economics, and parameter files.

Standard Coals--This feature allows the user to select from a menu of six representative coals in the event that the user cannot supply his/her own coal characteristics. The standard coals file contains the proximate analyses for run-of-mine (ROM) and physically cleaned coals. Weight recovery, Btu recovery, and total cost (in \$/ton of raw coal) are also shown for the cleaned coal.

Emission Calculations--The EPA AP-42 emission factors<sup>(5)</sup> used as a basis for these calculations are responsive to boiler bottom type (wet or dry) and coal type (rank) for PM; coal type for SO<sub>2</sub>; and firing configuration, bottom type, and coal type for NO<sub>x</sub>. For SO<sub>2</sub> emission calculations, the AP-42 basis is used; however, the user can select a separate default value percent conversion of sulfur



plants throughout the U.S. Selected results from these efforts, as they relate to integrated modeling and benchmarking FGD technology costs, are presented in the following section.

## RESULTS AND DISCUSSION

One method of evaluating cost estimates prepared for retrofit control systems is to benchmark them against actual retrofit control systems that are comparable in terms of design, performance, and operating characteristics. For this exercise, we selected FGD (wet and dry) as the technology of choice for the following reasons:

- FGD is the most commercially mature technology in the technology slate.
- FGD has been applied commercially as a retrofit control technology.
- The FGD modules (lime spray drying and wet lime/limestone slurry) in the IAPCS model represent the most versatile modules in terms of process design options and nuances that can be modeled.

Using the Flue Gas Desulfurization Information System (FGDIS)<sup>(6)</sup> seven retrofit FGD systems were selected for benchmarking. They are:

- Mitchell 33 of West Penn Power (lime slurry)
- Paradise 1 and 2 of TVA (limestone slurry)
- Four Corners 4 and 5 of Arizona Public Service (lime slurry)
- Wyodak 1 of Pacific Power and Light (dry lime)
- Riverside 6 and 7 of Northern States Power (dry lime)

Process design and operating data for these systems were extracted from FGDIS. Capital and annual cost data were extracted from the Form EIA 767 (Steam Electric Plant Operation and Design Report) for the reporting year of 1985. A summary of the pertinent design and operating characteristics for these systems is presented in Table 15. A comparison of the reported (actual) and model cost estimates is presented in Table 16.

Variations in the reported and model-generated cost estimates can be attributed to the following factors:

- Assignment of retrofit difficulty factors. Retrofit difficulty factors represent "adjustments" that allow new plant costs to be applied to existing plants in order to generate retrofit cost estimates. The RDF's used by PEI in acid rain control studies come from two sources. The first source represents site-specific RDF's that were developed from plant visits and detailed site investigations. The second source represents generic RDF's that were developed by PEI for use on plants in the absence of site-specific plant layout information. Five technology retrofit difficulty factor schedules were developed by PEI for these evaluations. Each technology category has been converted into a schedule which shows sensitivity to boiler size and age. It has been PEI's experience that RDF is primarily driven by these two parameters (i.e., smaller boilers tend to be more difficult to retrofit because of site congestion and older boilers tend to be more difficult to retrofit because of the "accumulation" of equipment over time). Our experience has shown that the variation in RDF's is a systematic function of these parameters. Using the base values and assigning these values to a relatively new (i.e., less than 10 years old) 500-MW unit and varying these values systematically as a function of size and age yields the technology RDF schedule.

Table 17 provides a description of the technology categories complete with the specific technologies contained in each category and Table 18 presents the add on technology RDF schedule.



- Assignment of indirect capital cost factors. The indirect capital costs and their accounting may vary with the modeling procedure. The following values were assigned for the cost model runs:

| <u>Indirect component</u>           | <u>LSD</u> | <u>FGD</u> |
|-------------------------------------|------------|------------|
| General facilities                  | 10         | 10         |
| Engineering and home office fees, % | 10         | 10         |
| Project contingency, %              | 30         | 30         |
| Process contingency, %              | 4.3        | 1.4        |
| Preproduction costs                 | Y          | Y          |
| Inventory capital                   | Y          | Y          |
| Idealized construction period, yr   | 2          | 3          |

- Assignment of unit operating costs. The unit costs associated with the FGD systems and the values used for modeling may vary. The following values were assigned for the cost model runs:

| <u>Item</u>          | <u>Value<br/>(1/85 \$)</u> | <u>Units</u>       |
|----------------------|----------------------------|--------------------|
| Operating labor      | 19.70                      | \$/person hour     |
| Water (river)        | 0.60                       | \$/1000 gal        |
| Lime                 | 65                         | \$/ton             |
| Limestone            | 15                         | \$/ton             |
| Land                 | 6,500                      | \$/acre            |
| Sludge, dry basis    | 9.75                       | \$/ton (dry basis) |
| Dry, granular solids | 8.00                       | \$/ton             |
| Electric power       | 5.0                        | c/kWh              |
| Steam                | 2.85                       | \$/1000 lb         |

- Determination of scope of supply. The scope of supply associated with the reported costs may vary from that assumed for modeling.

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## DISCLAIMER

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

TABLE 1. INTEGRATED MODELING PROJECTS

| Project       | Description                                                                                                                                                                   |
|---------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Ohio STAR     | Detailed retrofit control technology cost and performance estimates for five existing Ohio coal-fired utility power plants.                                                   |
| Ohio OCDO     | Retrofit control technology cost performance estimates for all existing Ohio coal-fired utility power plants.                                                                 |
| Ohio OAQDA    | Retrofit control and repowering technology cost and performance estimates for all existing Ohio coal-fired utility power plants.                                              |
| Kentucky STAR | Retrofit control technology cost and performance estimates for all stationary sources and detailed cost and performance estimates for seven existing coal-fired power plants. |
| NESCAUM STAR  | Control technology cost and performance estimates for all stationary and mobile sources in the northeastern U.S.                                                              |
| WESTAR        | Control technology cost and performance estimates for all stationary and mobile sources in the western U.S.                                                                   |
| NAPAP         | Detailed retrofit control technology cost and performance estimates for 12 mideastern plants and 50 plants in the top 200 population.                                         |
| GRI           | Control technology cost and performance estimates for the top 100 SO <sub>2</sub> -emitting plants in the eastern U.S.                                                        |

TABLE 2. COALBASE DATA ELEMENTS

| Descriptive               | Fuel                                 |
|---------------------------|--------------------------------------|
| Oris code                 | Coal rank                            |
| Utility                   | Heat content                         |
| Plant                     | Sulfur content                       |
| State                     | Pyritic sulfur                       |
| Unit No.                  | Ash percent                          |
| Boiler No.                | Moisture percent                     |
| Service date              | Volatile matter percent              |
| Status                    | Fixed carbon percent                 |
| Geographical regions      |                                      |
| Boiler                    | Emission Controls                    |
| Nameplate rating          | SO <sub>2</sub> standard             |
| Net dependable capability | NO <sub>x</sub> standard             |
| Boiler supplier           | PM standard                          |
| Draft type                | PM collector service date            |
| Firing type               | PM collection application            |
| Firing configuration      | PM type                              |
| Bottom type               | PM design                            |
| Furnace type              | PM removal                           |
| Furnace depth             | ESP collection area                  |
| Furnace width             | Fabric filter AC ratio               |
| Furnace height            | SO <sub>2</sub> control service date |
| Boiler roof-nose          | SO <sub>2</sub> control type         |
| Boiler bottom-nose        | SO <sub>2</sub> control design       |
| Furnace plan area         | SO <sub>2</sub> emission rate        |
| Multiple fuel             | NO <sub>x</sub> control type         |
| Gas availability          | NO <sub>x</sub> control design       |
| Coal rate                 | NO <sub>x</sub> emission rate        |
| Coal consumption          | Acid emissions                       |
| Heat rate                 | Acid emission rate                   |
| Capacity factor           |                                      |
| Load profile              |                                      |
| Heat input                |                                      |
| Steam rate                |                                      |
| Steam temperature         |                                      |
| Steam pressure            |                                      |
| Steam cycle               |                                      |
| Gas flow rate             |                                      |
| Gas temperature           |                                      |



TABLE 3. IAPCS CONTROL MODULES

| Category                  | Technology                                   | Process                                                                                               |
|---------------------------|----------------------------------------------|-------------------------------------------------------------------------------------------------------|
| Pre-combustion<br>In situ | Physical coal cleaning (PCC)                 | Conventional                                                                                          |
|                           | Low-NO <sub>x</sub> combustion (LNC)         | Low excess air<br>Overfire air<br>Low-NO <sub>x</sub> burner<br>Low-NO <sub>x</sub> concentric firing |
| Post-combustion           | Limestone injection multistage burner (LIMB) | Furnace sorbent injection                                                                             |
|                           | Reburning                                    | Natural gas                                                                                           |
|                           | Selective catalytic reduction (SCR)          | Hot-side                                                                                              |
|                           | Gas conditioning (GC)                        | SO <sub>3</sub><br>Spray humidification                                                               |
|                           | Dry sorbent injection (DSI)                  | Nahcolite<br>Trona                                                                                    |
|                           | Lime spray drying (LSD)                      | Spray dryer absorber<br>Duct spray drying                                                             |
|                           | Electrostatic precipitator (ESP)             | Cold-side                                                                                             |
|                           | Fabric filter (FF)                           | Reverse-air                                                                                           |
|                           | Flue gas desulfurization (FGD)               | Lime/limestone                                                                                        |

TABLE 4. COAL SUPPLY OPTIONS DESIGN AND PERFORMANCE PARAMETERS

|                             |                                                                               |
|-----------------------------|-------------------------------------------------------------------------------|
| Plant application           | New/retrofit                                                                  |
| Boiler application          | All firing types                                                              |
| Plant size, MW              | 100 to 1300                                                                   |
| Process options             | PCC<br>CS/B                                                                   |
| Coal selection options      | User specified--ROM<br>User specified--ROM/cleaned<br>Typical--model supplied |
| Coal quality                | ROM/cleaned                                                                   |
| Fuel price premium, \$/ton  | User specified/model calculated                                               |
| Coal cleaning cost benefits | Calculated                                                                    |
| Economic conditions         | TVA premises<br>EPRI premises                                                 |

TABLE 5. LNC DESIGN AND PERFORMANCE PARAMETERS

|                                       |                                                                                                       |
|---------------------------------------|-------------------------------------------------------------------------------------------------------|
| Plant application                     | New/retrofit                                                                                          |
| Boiler application                    | Pulverized coal                                                                                       |
| Boiler firing configuration           | Wall-fired and tangentially-fired                                                                     |
| Plant size, MW                        | 100 to 1300                                                                                           |
| Process options                       | Low-NO <sub>x</sub> burner<br>Overfire air<br>Low excess air<br>Low-NO <sub>x</sub> concentric firing |
| NO <sub>x</sub> control, percent      |                                                                                                       |
| Overfire air                          | 25 (default)                                                                                          |
| Low-NO <sub>x</sub> burner            | 50 (default)                                                                                          |
| Low excess air                        | 15 (default)                                                                                          |
| Low-NO <sub>x</sub> concentric firing | 40 (default)                                                                                          |
| Economic conditions                   | TVA premises<br>EPRI premises                                                                         |

TABLE 6. LIMB DESIGN AND PERFORMANCE PARAMETERS

|                             |                                         |
|-----------------------------|-----------------------------------------|
| Boiler application          | New/retrofit                            |
| Boiler firing configuration | All firing types                        |
| Plant size, MW              | 100 to 1300                             |
| Sorbent                     | Calcitic hydrate                        |
| Sorbent Ca/S Ratio          | Specified by user (default = 2)         |
| Sorbent injection option    | Upper-furnace injection                 |
| Process areas               |                                         |
| Sorbent handling            |                                         |
| Sorbent injection           |                                         |
| Boiler modifications        |                                         |
| Downstream modifications    | ESP upgrade (retrofit)                  |
| Waste handling and disposal | Gas conditioning (gas conditioning)     |
| Economic conditions         | System<br>TVA premises<br>EPRI premises |

TABLE 7. NGR DESIGN AND PERFORMANCE PARAMETERS

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|                                            |                                    |
|--------------------------------------------|------------------------------------|
| Boiler application                         | New/retrofit                       |
| Boiler firing configuration                | All firing types                   |
| Plant size, MW                             | 100 to 1300                        |
| Process options                            | Natural gas substitution           |
| Substitution, percent                      | 0 to 25 (default = 15)             |
| NO <sub>x</sub> removal, percent           | 0 to 60 (default = 60)             |
| Process areas                              |                                    |
| Natural gas service                        | Optional                           |
| Boiler modifications                       | Retrofit                           |
| Combustion modifications                   | Retrofit                           |
| Fuel injection system                      | New/retrofit                       |
| Fuel price premium, \$/10 <sup>6</sup> Btu | Specified by user (default = 2.00) |
| Economic conditions                        | TVA premises                       |
|                                            | EPRI premises                      |

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TABLE 8. SCR DESIGN AND PERFORMANCE PARAMETERS

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|                                                       |                                                             |
|-------------------------------------------------------|-------------------------------------------------------------|
| Boiler application                                    | New/retrofit                                                |
| Boiler firing configuration                           | All firing types                                            |
| Plant size, MW                                        | 100 to 1300                                                 |
| Process options                                       | Hot-side (high dust loading)                                |
| NH <sub>3</sub> :NO <sub>x</sub> stoichiometric ratio | Specified by user (default = 0.87)                          |
| NO <sub>x</sub> removal, percent                      | Specified by stoichiometric ratio<br>setting (default = 80) |
| Process areas                                         |                                                             |
| Ammonia storage and injection                         |                                                             |
| Reactors                                              |                                                             |
| Flue gas handling                                     |                                                             |
| Air heater modifications                              |                                                             |
| Catalyst life, yr                                     | Specified by user (default = 3)                             |
| Economic conditions                                   | TVA premises                                                |
|                                                       | EPRI premises                                               |

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TABLE 9. GC DESIGN AND PERFORMANCE PARAMETERS

|                                             |                              |
|---------------------------------------------|------------------------------|
| Boiler application                          | New/retrofit                 |
| Boiler firing configuration                 | All firing types             |
| Plant size, MW                              | 100 to 1300                  |
| Process options                             | Spray humidification         |
| Process options                             | SO <sub>3</sub> conditioning |
| Spray humidification                        |                              |
| Design                                      | Humidification chamber       |
| Saturation approach temperature, °F         | 160 (default)                |
| Chamber pressure drop, in. H <sub>2</sub> O | 1 (default)                  |
| Gas conditioning                            |                              |
| Design                                      | Catalytic combustion chamber |
| Feedstock                                   | Sulfur                       |
| Residual SO <sub>3</sub> , ppm              | 5 (default)                  |
| Economic conditions                         | TVA premises                 |
|                                             | EPRI premises                |

TABLE 10. DSI DESIGN AND PERFORMANCE PARAMETERS

|                                   |                                    |
|-----------------------------------|------------------------------------|
| Boiler application                | New/retrofit                       |
| Boiler firing configuration       | All firing types                   |
| Coal application                  | Low-sulfur coal                    |
| Plant size, MW                    | 100 to 1300                        |
| Process options                   | Nahcolite                          |
|                                   | Trona                              |
| Process design                    | Continuous-injection FF collection |
| SO <sub>2</sub> capture, percent  | 80 (default)                       |
| NO <sub>x</sub> capture, percent  | 22 (default)                       |
| Reagent stoichiometric ratio, NSR |                                    |
| Nahcolite                         | 0.75                               |
| Trona                             | 1.25                               |
| Economic conditions               | TVA premises                       |
|                                   | EPRI premises                      |



TABLE 11. LSD DESIGN AND PERFORMANCE PARAMETERS

|                                                      |                      |
|------------------------------------------------------|----------------------|
| Boiler application                                   | New/retrofit         |
| Boiler firing configuration                          | All firing types     |
| Plant size, MW                                       | 100 to 1300          |
| Process options                                      | Spray dryer absorber |
|                                                      | Duct spray drying    |
| Process design                                       | ESP upgrade          |
|                                                      | Replacement FF       |
| SO <sub>2</sub> removal efficiency, maximum percent  | 73 to 86 (SDA)       |
|                                                      | 40 to 60 (DSD)       |
| SO <sub>2</sub> removal across PM collector, percent |                      |
| ESP                                                  | 21 (default)         |
| FF                                                   | 55 (default)         |
| Saturation approach temperature, °F                  | 20 (default)         |
| Reagent stoichiometric ratio, equivalent Ca/S        | 1.4 (default)        |
| Slurry recycle fraction, percent                     | 50 (default)         |
| Slurry recycle solids, percent by weight             | 35 (default)         |
| Lime preparation                                     | Ball mill/slaker     |
| Economic conditions                                  | TVA premises         |
|                                                      | EPRI premises        |

TABLE 12. ESP DESIGN AND PERFORMANCE PARAMETERS

|                                   |                           |
|-----------------------------------|---------------------------|
| Plant application                 | New/retrofit              |
| Boiler application                | All firing types          |
| Coal application                  | All coal types            |
| Process options                   | Cold-side                 |
| Process design                    | Rigid frame               |
| Process areas                     |                           |
| ESP box                           |                           |
| Ductwork                          |                           |
| Dry ash handling                  |                           |
| Process features                  |                           |
| Gas conditioning                  | GC module                 |
| Additional plate area             | ESP upgrade               |
| Additional insulation             | LSD module                |
| PM collection efficiency, percent | Calculated/user specified |
| Economic conditions               | TVA premises              |
|                                   | EPRI premises             |

TABLE 13. FABRIC FILTER DESIGN AND PERFORMANCE PARAMETERS

|                                   |                  |
|-----------------------------------|------------------|
| Plant application                 | New/retrofit     |
| Boiler application                | All firing types |
| Coal application                  | All coal types   |
| Process design                    | Reverse air      |
| Process areas                     |                  |
| Baghouse                          |                  |
| Self-cleaning system              |                  |
| Ductwork                          |                  |
| Dry ash handling                  |                  |
| PM collection efficiency, percent | 99.7 (maximum)   |
| Air/cloth ratio                   | 2:1 (default)    |
| Economic conditions               | TVA premises     |
|                                   | EPRI premises    |

TABLE 14. FGD DESIGN AND PERFORMANCE PARAMETERS

|                                  |                           |
|----------------------------------|---------------------------|
| Design parameters                |                           |
| Plant application                | New/retrofit              |
| Plant size, MW                   | 100 to 1300               |
| Number of absorbers              | 0 to 10                   |
| Number of spare absorbers        | 0 to 2                    |
| SO <sub>2</sub> removal, percent | 0 to 100                  |
| Process areas                    |                           |
| FGD                              | Raw material handling     |
|                                  | Raw material preparation  |
|                                  | SO <sub>2</sub> scrubbing |
|                                  | Oxidation                 |
|                                  | Reheat                    |
| System                           | Gas handling              |
|                                  | Waste disposal            |
| Process options                  | Limestone slurry          |
|                                  | Lime slurry               |
|                                  | Promoted lime/limestone   |
| Absorber options                 | Spray tower               |
|                                  | Packed                    |
|                                  | Venturi-spray tower       |
| Reheat options                   | Indirect steam            |
|                                  | Flue gas bypass           |
| Solid waste treatment options    | Chemical fixation         |
|                                  | Forced oxidation          |
| Solid waste disposal options     | Onsite/offsite            |
| Economic conditions              | TVA premises              |
|                                  | EPRI premises             |

TABLE 15. RETROFIT FGD SYSTEMS DESIGN AND OPERATING CHARACTERISTICS

|                            | Four<br>Corners | Four<br>Corners | Riverside | Wyodak | Paradise  | Paradise  | Mitchell |
|----------------------------|-----------------|-----------------|-----------|--------|-----------|-----------|----------|
| Unit                       | 4               | 4               | 6-7       | 91     | 1         | 2         | 3        |
| Size, MW                   | 739             | 739             | 140       | 362    | 663       | 663       | 284      |
| Firing method              | PC              | PC              | PC        | PC     | C         | C         | PC       |
| Boiler bottom              | Dry             | Dry             | Dry       | Dry    | Wet       | Wet       | Dry      |
| Capacity factor, %         | 65              | 65              | 65        | 65     | 65        | 65        | 65       |
| Coal, Btu/lb               | 8772            | 8772            | 8934      | 8090   | 11,644    | 11,657    | 12,227   |
| Coal sulfur, %             | 0.8             | 0.8             | 12        | 0.6    | 2.9       | 2.9       | 2.5      |
| Coal ash, %                | 22.8            | 22.8            | 92        | 6.7    | 8.5       | 8.5       | 13.2     |
| Gas flow rate, Macfm       | 2,858           | 2,858           | 640       | 2,352  | 2,112     | 2,112     | 1,100    |
| Gas temperature, °F        | 285             | 285             | 350       | 364    | 300       | 300       | 300      |
| Process                    | Lime            | Lime            | LSD       | LSD    | Limestone | Limestone | Mg-lime  |
| SO <sub>2</sub> removal, % | 72              | 72              | 70        | 75     | 84        | 84        | 95       |

TABLE 16. REPORTED AND ESTIMATED RETROFIT FGD COSTS

|            | Four<br>Corners | Four<br>Corners | Riverside | Wyodak | Paradise | Paradise | Mitchell |
|------------|-----------------|-----------------|-----------|--------|----------|----------|----------|
| Reported   |                 |                 |           |        |          |          |          |
| Capital    |                 |                 |           |        |          |          |          |
| \$MM       | 282.7           | 282.7           | 34.7      | 64.6   | 150.6    | 150.6    | 102.7    |
| \$/kW      | 382.5           | 382.5           | 247.8     | 178.5  | 227.1    | 227.1    | 361.6    |
| Annual O&M |                 |                 |           |        |          |          |          |
| \$MM       | 3.7             | 3.7             | 0.8       | NR     | 4.7      | 3.2      | 4.3      |
| Mills/kWh  | 1.49            | 1.09            | 1.35      | NR     | 3.23     | 1.13     | 3.92     |
| Estimated  |                 |                 |           |        |          |          |          |
| Capital    |                 |                 |           |        |          |          |          |
| \$MM       | 134.0           | 128.3           | 60.5      | 81.6   | 151.4    | 151.3    | 109.3    |
| \$/kW      | 181.4           | 173.6           | 432.0     | 225.4  | 228.4    | 228.2    | 365.4    |
| Annual O&M |                 |                 |           |        |          |          |          |
| \$MM       | 22.6            | 22.1            | 6.2       | 9.0    | 27.8     | 27.8     | 18.3     |
| Mills/kWh  | 5.4             | 5.3             | 7.7       | 4.4    | 7.4      | 7.4      | 10.8     |

TABLE 17. DESCRIPTION OF TECHNOLOGY CATEGORIES

| Technology category | Affected boiler area                                          | Description                                                                                                                                                                                                                                                                             | Specific technologies    |
|---------------------|---------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------|
| Add on              | Between PM collector and stack                                | Add on to tail end of the boiler and require no boiler modifications. RDF accounts for need to fit equipment into available space.                                                                                                                                                      | FGD<br>SCR               |
| Cold duct           | Between air preheater and PM collector                        | Insert between the discharge of the air preheater and the inlet of the PM collector. RDF accounts for minor modifications to the boiler and ductwork and the need to fit equipment into available space.                                                                                | LSD<br>DSI               |
| Hot duct            | Between economizer and air preheater                          | Insert between the discharge of the inlet of the air preheater. RDF accounts for extensive modifications to the economizer, air preheater, and associated ductwork and the need to fit equipment into available space.                                                                  | SNR<br>SCR               |
| In situ             | Furnace and radiant and convective superheaters and reheaters | Incorporate into the furnace extending through the radiant and convective superheat and reheat recovery systems. RDF accounts for extensive modification to the burners, furnace, and superheater and reheater assemblies and the need to fit auxiliary equipment into available space. | LIMB<br>LNC<br>NGR<br>SC |
| Pre-combustion      | Fuel storage, preparation, handling, and conversion           | Add to the front end of the boiler and require no boiler or ductwork modifications. RDF accounts for modifications to fuel storage, handling, and preparation equipment and the need to fit fuel conversion equipment into available space.                                             | PCC                      |

TABLE 18. ADD-ON TECHNOLOGY RDF SCHEDULE

| Age, yr | Size, MW |         |         |         |         |          |       |
|---------|----------|---------|---------|---------|---------|----------|-------|
|         | <100     | 100-300 | 300-500 | 500-700 | 700-900 | 900-1100 | >1100 |
| <10     | 1.45     | 1.40    | 1.35    | 1.30    | 1.25    | 1.20     | 1.15  |
| 10-20   | 1.50     | 1.45    | 1.40    | 1.35    | 1.30    | 1.25     | 1.20  |
| 20-30   | 1.55     | 1.50    | 1.45    | 1.40    | 1.35    | 1.30     | 1.25  |
| >30     | 1.60     | 1.55    | 1.50    | 1.45    | 1.40    | 1.35     | 1.30  |



## SITE-SPECIFIC EVALUATION OF SIX SORBENT

### INJECTION PROCESSES

By: P. A. Ireland and G. D. Brown  
United Engineers and Constructors, Inc.  
Stearns-Roger Division  
700 South Ash Street  
P.O. Box 5888  
Denver, CO 80217  
and  
W. F. Frazier  
Virginia Power  
5000 Dominion Blvd.  
Glen Allen, Virginia 23060  
and  
P. T. Radcliffe  
Electric Power Research Institute  
3412 Hillview Avenue  
P.O. Box 10412  
Palo Alto, CA 94303

### ABSTRACT

There is currently much interest and development work occurring in the area of sorbent injection as a retrofit technique for SO<sub>2</sub> control for coal-fired power plants. This paper presents results from a recent study which evaluated the technical feasibility and developed site-specific costs for the following six sorbent injection processes.

- ° Furnace Sorbent Injection
- ° Duct Sorbent Injection
- ° Duct Spray Drying
- ° Lime Spray Drying
- ° Lurgi CFB Absorption
- ° Tampella LIFAC

Comparative costs are presented for the six processes based on design and performance data supplied by vendors, published literature, and process developers. Site-specific retrofit costs were developed for each technically feasible application of each process on five Virginia Power coal-fired generating units located at two different power stations which burn a 1.0 wt% sulfur (ave.) Central Appalachian bituminous coal. Levelized costs are presented in \$/ton of SO<sub>2</sub> removed to account for different process performances and for comparison to other technologies. Sensitivities are presented on several variables to show applicability to other conditions.

## INTRODUCTION

Developing dry sorbent SO<sub>2</sub> control processes have the potential to be more cost effective than either wet scrubbers or fuel switching as a means of reducing SO<sub>2</sub> emissions from coal-fired generating units, particularly for moderate removal requirements<sup>(1)</sup>. At the Chesterfield and Possum Point Stations owned by Virginia Power, space for retrofitting wet scrubbers is severely limited - the electrostatic precipitators have been built virtually on the river banks located behind the generating units.

Virginia Power and the Electric Power Research Institute co-sponsored a site-specific evaluation of the following six different SO<sub>2</sub> emission control technologies at the Chesterfield and Possum Point Power Stations:

- (1) Furnace Sorbent Injection (FSI)
- (2) Duct Sorbent Injection (DSI)
- (3) Duct Spray Drying (DSD)
- (4) Lime Spray Drying (Spray Dryer)
- (5) Lurgi Circulating Fluid Bed Absorption (Lurgi)
- (6) Tampella LIFAC (Tampella)

The objective of this study was to determine the technical feasibility and economics of applying these six sorbent injection schemes to each of the coal-fired generating units located at the Chesterfield and Possum Point Power Stations. As part of this study, sensitivity analyses were performed to determine the major variables affecting the emission control costs. This paper presents results from this study.

## DESIGN CRITERIA AND ASSUMPTIONS

An economic analysis was performed based on technical and economic criteria specific to the Virginia Power Chesterfield and Possum Point Stations<sup>(2)</sup>. These two stations, both located in Virginia, burn a low to medium sulfur coal from the Central Appalachia region of eastern Kentucky, southern West Virginia and southwestern Virginia. The capital costs for the processes were based on a 1.5 wt percent sulfur coal (HHV = 12,000 Btu/lb, Ash = 15 wt% and moisture = 8 wt%) and the operating costs were based on a more typically expected 1.0 wt percent sulfur coal (HHV = 12,500 Btu/lb, Ash = 10 wt% and Moisture = 6 wt%). The generating units at these stations were built when scrubbers were not required, and as such, there is no room for scrubbers behind the existing electrostatic precipitators. The back of the precipitators are located along the bank of the James and Potomac Rivers at the Chesterfield and Possum Point Power Stations, respectively. At both stations, flue gas desulfurization (FGD) absorbers would have to be located at "remote" locations to the side of the existing buildings, resulting in long and costly interconnecting ductwork.

The capital costs were based on vendor quotations for process equipment. Sizing information for the absorber/reactor vessels was obtained from the process vendors. In order to develop an accurate, credible cost estimate for the Spray Dryer, Lurgi and Tampella processes, which require the addition of an absorber/reactor vessel, a site-specific structural and foundation design and material takeoff for the vessels was developed for Chesterfield 4 and a detailed cost estimate made for this unit for each of these three processes. The cost accounts such as piping, electrical and instrumentation were factored specifically for each process. The standard EPRI format for capital cost estimates was used <sup>(3)</sup>, which includes costs for general facilities, engineering and home office fees, process contingency and project contingency.

The economic analysis used in this study assumed that all six of the processes would require the demolition of the existing wet sluice system for ash removal and the addition of a new dry ash pneumatic conveying waste disposal system because of the presence of unreacted quicklime or hydrated lime in the spent sorbent/fly ash waste mixture, which could potentially plug the system due to a pozzolanic reaction. Most of the current domestic sorbent injection process developers and utilities testing this technology were contacted and were found to be using a dry pneumatic conveying system to avoid this problem. However, future development may show this assumption to be incorrect, and allow wet sluicing of hydrated lime waste to disposal, thus reducing the cost of these systems.

The precipitators for all of the generating units were designed for low sulfur coal applications resulting in SCA's in the 400 to 600 range. With humidification of the flue gas included in each process design, the specific collection areas (SCA's) for the ESP's were judged to be adequate to maintain existing particulate emission limits based on limited modeling projections and actual field test data. Again, future development may show this assumption to be incorrect.

A listing of the technical and economic design basis used for this evaluation is shown in Table 1. The sorbent performance values used for each process are shown in Table 2, and in Figure 1 graphically. The performance assumptions shown in Table 2 for the FSI, DSI and DSD processes were based upon the results of pilot scale testing performed by EPRI and others.

The base case sorbent performance values chosen for each process were selected to provide the lowest levelized control cost for each process. Sorbent performance data was used in the computer economic model to determine the level of SO<sub>2</sub> removal which yielded the lowest levelized control cost for each process. The optimum SO<sub>2</sub> removal value for the Tampella process was not calculated due to a lack of sufficient available data. An example curve of control cost versus SO<sub>2</sub> removal for the FSI process is shown in Figure 2. An SO<sub>2</sub> removal of 40% was chosen for the FSI process which corresponds to its optimum (lowest cost) value. The graph shows decreasing control cost up to 40% SO<sub>2</sub> removal due to equipment cost economies of scale, and increasing control cost above 40% due to poorer sorbent utilization. All SO<sub>2</sub> removal processes exhibit this same trend.

#### PROCESS DESCRIPTIONS

The following discussion provides a brief description of the six processes evaluated.

1. Furnace Sorbent Injection (FSI) - Inject hydrated lime into furnace at 2200°F; humidification of flue gas to 200°F ahead of ESP for ESP conditioning only. Less expensive limestone can also be used as a sorbent, but economics show a preference for hydrated lime.
2. Duct Sorbent Injection (DSI) - Inject hydrated lime ahead of ESP; humidification of flue gas to 150°F just ahead of sorbent injection for both SO<sub>2</sub> removal and ESP conditioning.
3. Duct Spray Drying (DSD) - Inject lime slurry into duct ahead of ESP with simultaneous humidification of flue gas to 180°F. Atomizers may be either dual-fluid or rotary atomizers.

4. Lime Spray Drying (Spray Dryer) - Inject lime slurry into a Lime Spray Dryer (carbon steel construction) ahead of the ESP. Flue gas is cooled to 150°F. Some of the spent sorbent and fly ash captured in the ESP may be recycled back to the Lime Spray Dryer to increase lime utilization in the process. The spray dryer uses dual-fluid nozzles and is designed with a larger than normal height to diameter ratio in order to reduce the SO<sub>2</sub> absorber space requirements. The large spray dryer height to diameter ratio has been used for industrial units, but has not yet been demonstrated on a utility application.
5. Lurgi CFB Absorption (Lurgi) - Inject water and hydrated lime into a fluid bed absorber (carbon steel construction). Flue gas is cooled to 150°F. A portion of the entrained spent sorbent and fly ash is captured downstream in a mechanical dust collector and the existing ESP, and recirculated to the fluid bed absorber.
6. Tampella LIFAC (Tampella) - Inject limestone into furnace (at 2200°F) similar to FSI. Inject water into an "activation" reactor (316 SS construction) ahead of the existing ESP to humidify the flue gas to 140°F. About 20% of the flue gas downstream of the ESP is heated with steam and recycled to the outlet of the activation reactor to reheat the flue gas stream before entering the ESP.

Schematic diagrams of these six processes as evaluated are shown in Figures 3 and 4.

#### PROCESS APPLICABILITY

The six processes studied are currently at different levels of development and each process has advantages and disadvantages. A summary of the applicability of each process and its level of development is shown in Table 3.

The FSI, DSI and DSD processes are capable of being applied to a retrofit unit where no more than 40-50% (FSI and DSI) or 50-60% (DSD) SO<sub>2</sub> removal is required. These processes each require a minimum of one second of ductwork residence time between the airheater and ESP (or baghouse). All three of these processes also have a very low space requirement, and are, therefore, well suited for difficult retrofit situations. The FSI, DSI and DSD processes are currently being developed in the U.S. and Canada. The FSI process has been commercially applied in western Europe to boilers ranging in size from 15 to 700 MW. All of these European boilers burn low sulfur coals and typically have large ESP's. The DSI and DSD process performance is current based on pilot plant results. This study has assumed that any problems arising during demonstration of the processes will be successfully resolved.

The Spray Dryer and Lurgi processes can be applied to either new or retrofit units since they are capable of meeting NSPS. These two processes require adequate space between the airheater and ESP or baghouse to install one or more SO<sub>2</sub> absorbers. These processes are, therefore, best suited to new units or retrofit units which have this space available. The Spray Dryer process has been commercially applied to about 18 utility units in the U.S. ranging in size from 44 to 810 MW while the Lurgi process has been commercially applied to four utility units in West Germany ranging in size from 50 to 170 MW.



The Tampella process has achieved 80% SO<sub>2</sub> removal on European coals. This process has the advantage that it requires a limestone sorbent instead of the more expensive hydrated lime sorbent required by four of the other five processes. The Tampella process also requires adequate space between the airheater and ESP or baghouse to install one or more activation reactors. The Tampella process has been commercially applied to one 260 MW utility unit in Finland.

In all the processes studied, sorbent is added before the ESP thus causing a concern about excessive particulate emissions. Because of this ESP performance concern, all of these processes are best applied to a lower sulfur coal (requiring less sorbent) to minimize the risk of excess particulate emissions. In addition, the FSI, DSI and DSD processes are best applied on a low sulfur coal since they are "low capital/high operating" cost processes whose economics favor lower sulfur.

The Spray Dryer, Lurgi and Tampella processes are generally more economically attractive for a higher sulfur coal application since they are capital cost intensive and have a higher SO<sub>2</sub> removal and better sorbent utilization than the other three processes. They are also best applied to units with longer remaining service lives and higher capacity factors.

## RESULTS

### BASE CASE ECONOMIC RESULTS

The capital and levelized control cost results for the six processes applied to Chesterfield 4 are shown in Figures 5 and 6, respectively. The "base case" economic analysis for the six processes was performed for Chesterfield 4 since this unit is of medium size (166 MW) and has a plant life (18 years) that is typical of the five Virginia Power units examined. The results show that the DSI, DSD, Spray Dryer and Lurgi processes have equivalent levelized control costs within the accuracy of this study. The results also show that the FSI, DSI, and DSD processes, which do not require the addition of absorber vessels, are lower in capital cost than the Spray Dryer, Lurgi and Tampella processes which require the addition of absorber/ activation reactor vessels.

This study assumed that the FSI, DSI and DSD processes could be tied-in to the existing power plant during a normal 30 day outage. The Spray Dryer, Lurgi and Tampella processes, because of the need to retrofit an absorber vessel between the air heater and ESP on this particular unit, require a much longer equipment tie-in period for the vessels and, therefore, incur a replacement power cost penalty. The replacement power cost penalty comprises from 10 to 16 percent of the control cost of these three processes on Chesterfield 4. The capital costs for each process as presented in this paper are for equipment erection only, and do not include the replacement power cost penalty.

It is important to recognize that the results of the process screening evaluation are based on site-specific technical and economic information plus process performance assumptions (e.g. Ca/S molar ratio and SO<sub>2</sub> removal percentages) that remain to be proven at demonstration and/or full-scale applications for some of these processes. The relative ranking of the technology options at other sites could be influenced by major technical differences at the sites (e.g. presence of wet or dry ash removal systems, need for ESP upgrades, need for economizer replacements for FSI or Tampella, complexity of ductwork arrangements, space availability, ductwork residence time) and by differences in operating and cost assumptions (e.g. sorbent costs, unit capacity factors, design fuel characteristics, remaining plant life). On-going development work, plus

additional technology design optimization evaluations (e.g. use of recycle, sorbent promoters and use of flue gas reheat) may lead to changes in the process performance compared to that assumed in the subject study. Sensitivity analyses were performed to address the importance of these assumptions.

### **SENSITIVITY ANALYSES**

Through the use of a computer spreadsheet program, the cost impact of 14 technical and economic process variables was examined. The variables which were found to have a significant impact on the levelized control cost are as follows:

#### Major Impact (In Order of Importance)

- ° Plant Capacity Factor
- ° Sorbent Performance (FSI, DSI and DSD only)
- ° Levelized Fixed Charge Rate
- ° Ash Disposal Method (wet or dry)
- ° Capital cost of the absorber/reactor (Spray Dryer, Lurgi, and Tampella only)

#### Moderate Impact (In Order of Importance)

- ° Delivered sorbent cost
- ° Inflation rate

A sensitivity analysis of the six sorbent injection processes was performed to illustrate how the most important technical process uncertainties affect the relative costs. The six processes were compared at the base conditions on Chesterfield 4 and also at optimistic and pessimistic assumption cases for three process uncertainties. The optimistic case used favorable projected values for sorbent performance, reactor/absorber or humidification system capital cost and type of spent sorbent/fly ash disposal method which produced a low control cost. The pessimistic case used unfavorable projected values for these parameters which produced a high control cost.

The results of this analysis are shown in Table 4. The decrease in control cost from the base case to the optimistic case ranged from 31 to 36% for the FSI, DSI and DSD processes and from 17 to 18% for the Spray Dryer, Lurgi and Tampella processes. The increase in control cost for the pessimistic assumptions ranged from 21 to 31% for the FSI, DSI, and DSD processes and from 9 to 13% for the Spray Dryer, Lurgi and Tampella processes. These ranges show the larger uncertainty in cost associated with the FSI, DSI, and DSD processes which have not been developed much above the pilot stage and a moderate uncertainty in cost associated with the Spray Dryer, Lurgi and Tampella processes which have been demonstrated at a commercial level. A bar graph of the process control costs versus the design assumption cases is shown in Figure 7.

Based on this sensitivity analysis, it is difficult to make a clear economic choice between the DSI, DSD, Spray Dryer and Lurgi processes since over the range of costs examined there is considerable overlap in control costs for these four processes when compared at the same conditions. Further process development work is needed to more clearly define the costs of these four processes before a clear economic choice can be made. This sensitivity analysis is based only on economic factors and does not take the relative risks associated with each process into account.

The FSI and Tampella processes were somewhat higher in control cost than the other four processes as applied specifically to Chesterfield 4. The FSI process evaluated higher in cost because of its lower reagent utilization compared to the other five processes. The FSI process would be more applicable to units where there was inadequate space for absorber vessels, and where only partial flue gas humidification was possible.

The Tampella, Spray Dryer and Lurgi processes evaluated higher in capital cost because of the small space available for the absorber/reactor vessels (requiring two vessels for each process) and the high retrofit costs associated with this unit. In addition, the Tampella process, because of its closer approach to saturation temperature, also requires that the activation reactors be fabricated from 316 SS material, and includes flue gas reheat whereas reheat is not included in the other processes.

#### EXTRAPOLATION OF RESULTS TO OTHER UNITS

For each unit under study, the technical feasibility of each of the six alternative sorbent injection schemes was determined. This evaluation was based on critical process requirements and plant compatibility factors including available duct residence time, available SCA in the existing precipitator, on-site space availability, access for system construction and duct arrangement.

The criteria used in the evaluation of technical feasibility for each of the six processes were:

- ° FSI - All units should be capable of FSI retrofit unless evaluation of the boiler drawings indicated possible problems with obstructions in the furnace injection zone (all units were acceptable). Ductwork ahead of ESP must have at least one half second residence time (in straight section) for partial humidification (Chesterfield 6 was eliminated based on this criterion).
- ° DSI and DSD - All units should be capable of DSI and DSD sorbent injection retrofit unless the flue gas residence time between the air heater and new ESP is less than one second in straight lengths of ductwork (as with Chesterfield Unit 6).
- ° Spray Dryer, Lurgi and Tampella - These three processes all require the addition of reactor/absorber vessels between the existing airheaters and ESP. Therefore, space must be available in this area for these vessels and for access during construction. The Chesterfield Units 5 and 6 were eliminated based on their lack of available space for the reactor/absorber vessels and lack of construction access.

All the technically feasible sorbent injection options were further evaluated economically in the study.

The results of this analysis for the other units showed that the capital costs for the Spray Dryer, Lurgi and Tampella processes (\$85-220/KW) are generally lower in comparison to typical retrofit FGD costs of \$250-500/KW, but higher than the FSI, DSI and DSD retrofit capital costs of \$30-60/KW. The relative control cost ranking of the processes remained the same for each unit studied. The rankings did not change between the units because the retrofit difficulty, technical design criteria and economic factors used in the analysis were similar for all five of the units studied.



## CONCLUSIONS

Specific conclusions from the site-specific evaluation of six sorbent injection SO<sub>2</sub> control processes include the following:

1. All six of the processes were technically feasible at Possum Point 3 & 4 and Chesterfield 3 & 4. The FSI, DSI and DSD processes are also technically feasible at Chesterfield 5, but not at Chesterfield 6 because of insufficient duct residence time ahead of the existing ESP for sorbent injection and humidification. The Spray Dryer, Lurgi and Tampella processes are not technically feasible at Chesterfield 5 & 6 because of the lack of available space for the reactor/absorber vessels and lack of construction access.
2. The capital costs for the FSI, DSI and DSD processes were in the range of \$50 to \$58/KW while the Spray Dryer and Lurgi processes, which include the addition of SO<sub>2</sub> absorbers, ranged from \$97 to \$118/KW for Chesterfield Unit 4, the base case unit for this study.
3. The capital cost for the Tampella Process at \$176/KW is much higher than the capital cost of the Spray Dryer or Lurgi Processes primarily due to the activation reactor material being 316 SS and the addition of a recycle flue gas reheat system for Tampella.
4. The DSI, DSD, Spray Dryer and Lurgi processes have the lowest levelized control cost (\$/ton SO<sub>2</sub> removed) when applied to Chesterfield Unit 4 and are considered to be equivalent in control cost within the accuracy of this study.
5. The results of this study are entirely site-specific to the two Virginia Power Stations examined. Items applicable to other sites which could change the results of this analysis are:
  - ° More available space between the airheater and ESP to allow for the retrofit of a single SO<sub>2</sub> absorber instead of two absorbers for the Spray Dryer, Lurgi and Tampella processes.
  - ° Different reagent prices
  - ° No need for a new dry pneumatic waste disposal system
  - ° A high or medium sulfur coal application instead of the low sulfur coal used in this study. Coal sulfur has a major impact on the costs of the processes.
6. The six processes studied are currently at different levels of development. Therefore, the relative risk of each process becomes an important factor, along with project costs, in selecting a process.

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.



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2. Frazier, W.F. of Virginia Power, Ireland, P.A. and Brown, G.D. of Stearns-Roger Division of United Engineers and Constructors, Inc., and McElroy, M.W. of EPRI, "A Site-Specific Evaluation of Dry SO<sub>2</sub> Emission Controls", presented at the 1987 Joint Power Generation Conference, in Miami, Florida, October 1987.
3. Electric Power Research Institute (EPRI), Technical Assessment Guide, EPRI P-4463-SR, Volume 1, December, 1986.

## DISCLAIMER

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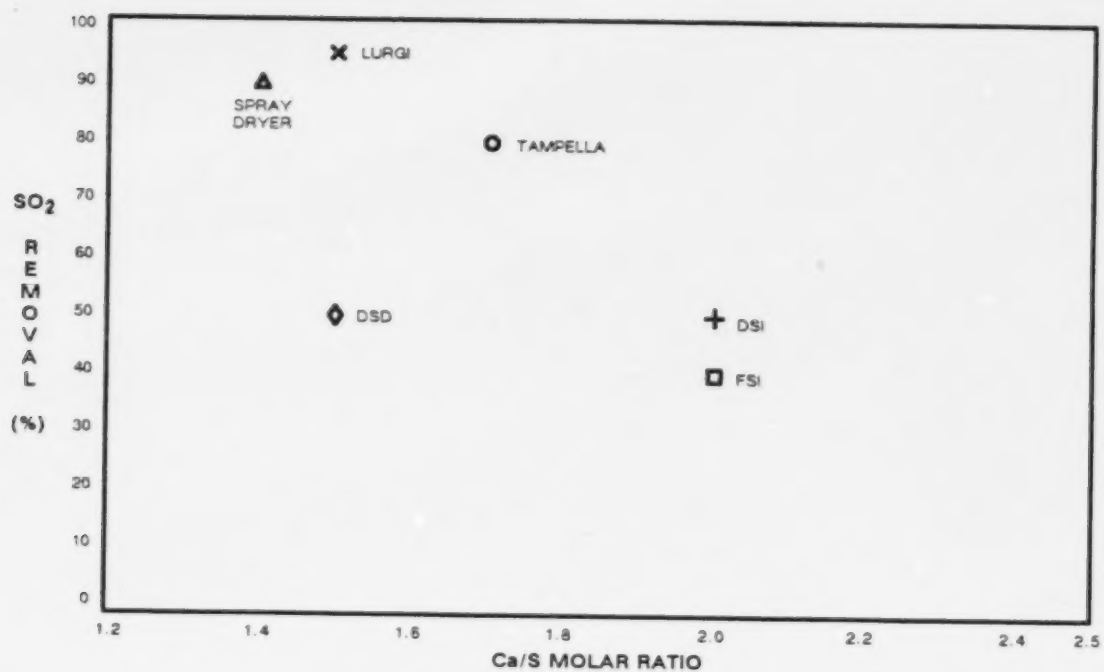


Figure 1. Operating Sorbent Performance  
Six Sorbent Injection Processes

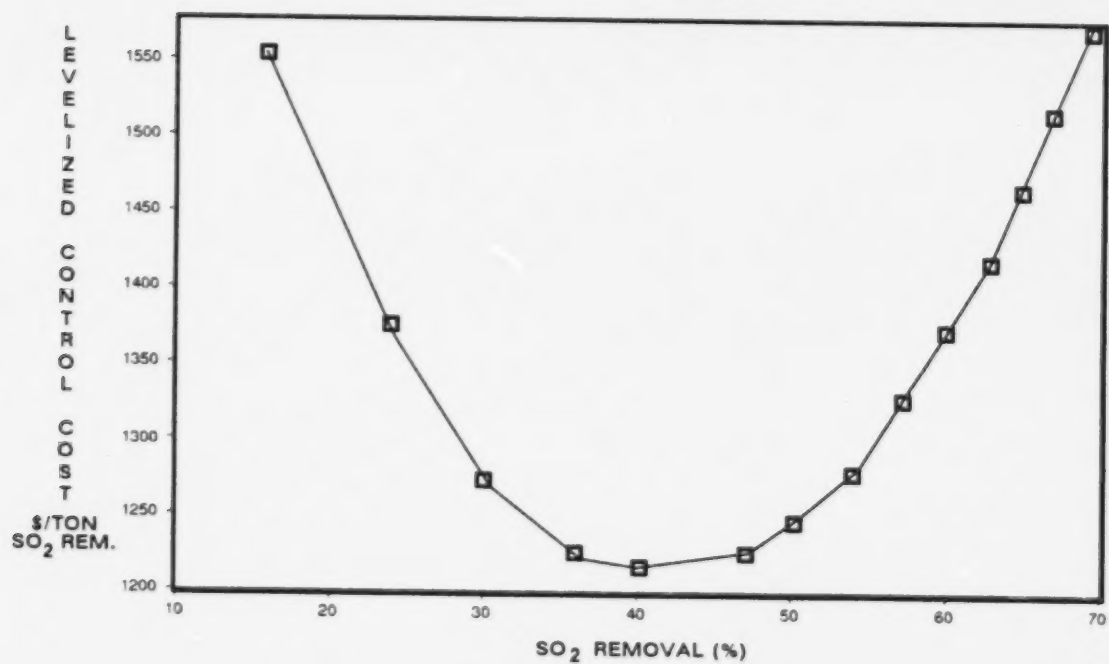
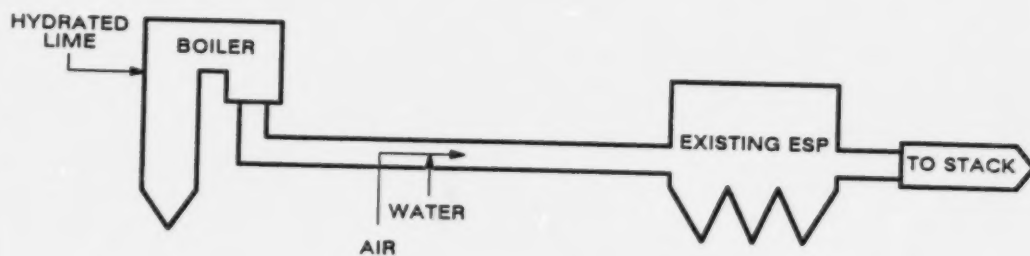
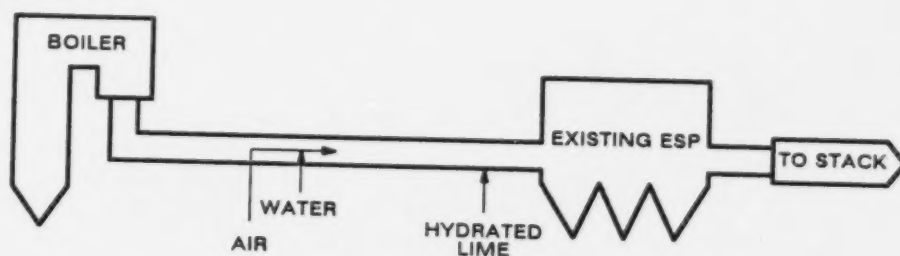


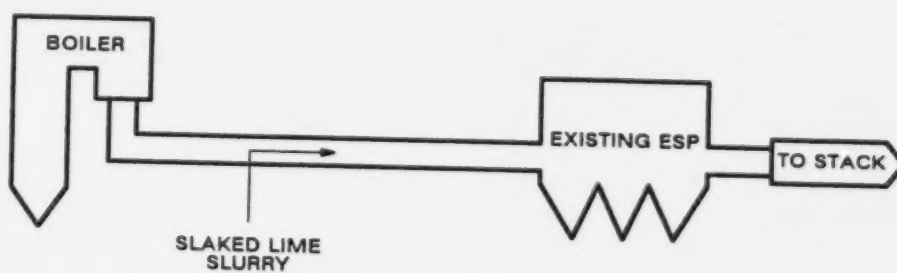
Figure 2. Control Cost vs SO<sub>2</sub> Removal  
Furnace Sorbent Injection Process



FURNACE SORBENT INJECTION



DUCT SORBENT INJECTION



DUCT SPRAY DRYING

Figure 3. Process Flow Diagrams  
for the FSI, DSI and DSD Processes

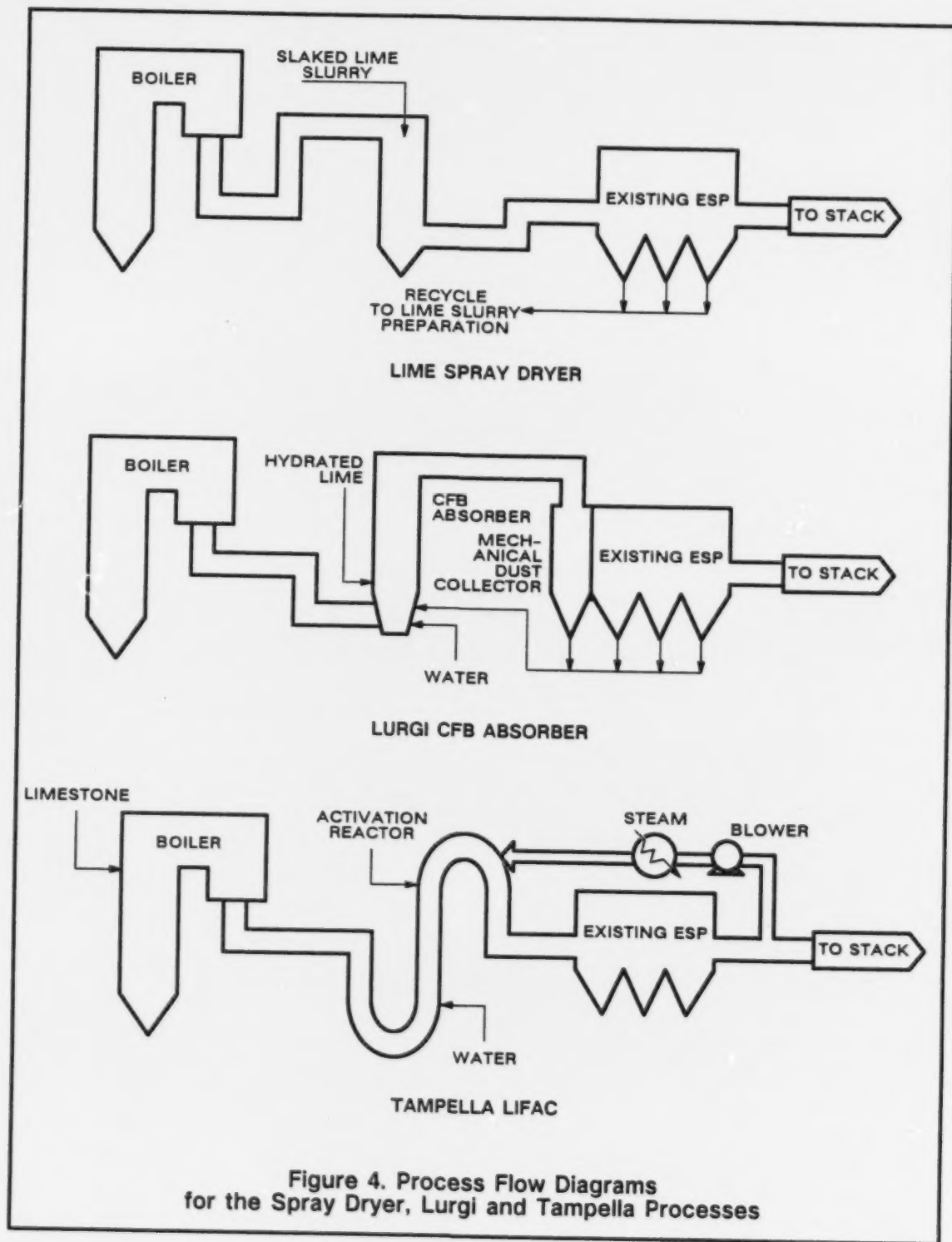


Figure 4. Process Flow Diagrams  
for the Spray Dryer, Lurgi and Tampella Processes



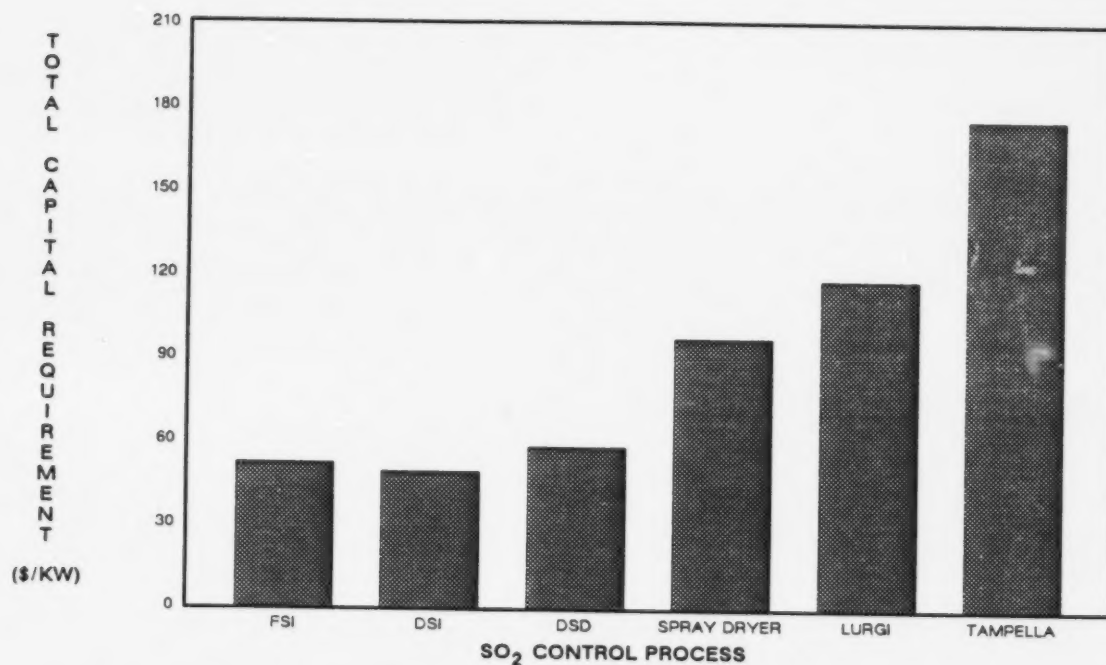


Figure 5. Total Capital Requirement  
Virginia Power/Chesterfield 4

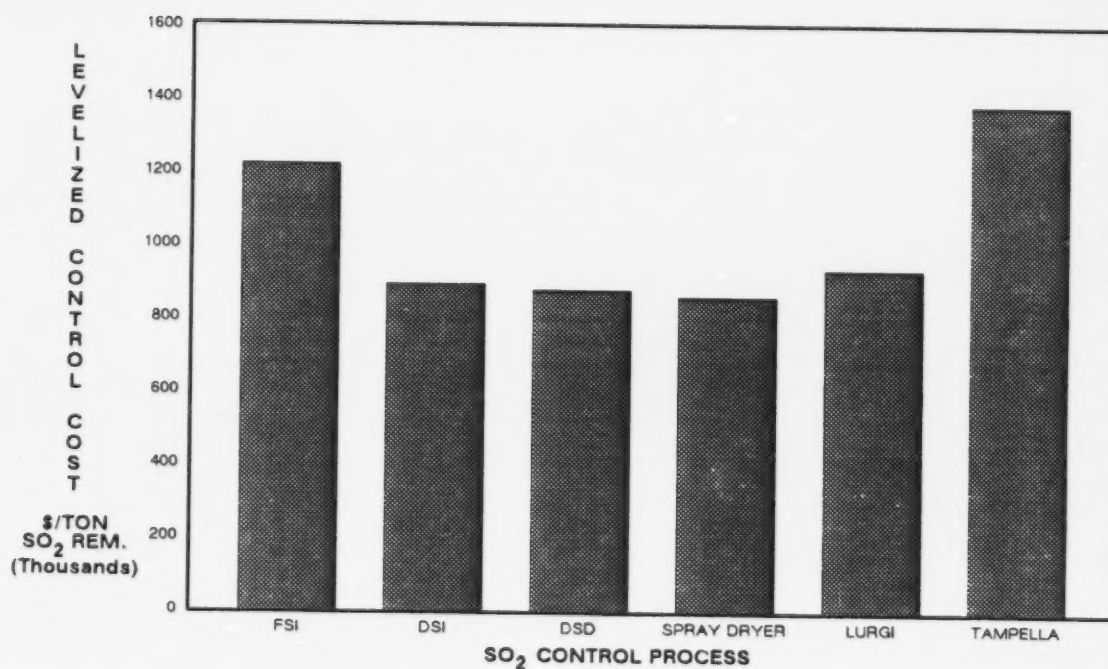


Figure 6. Levelized Control Cost  
Virginia Power/Chesterfield 4

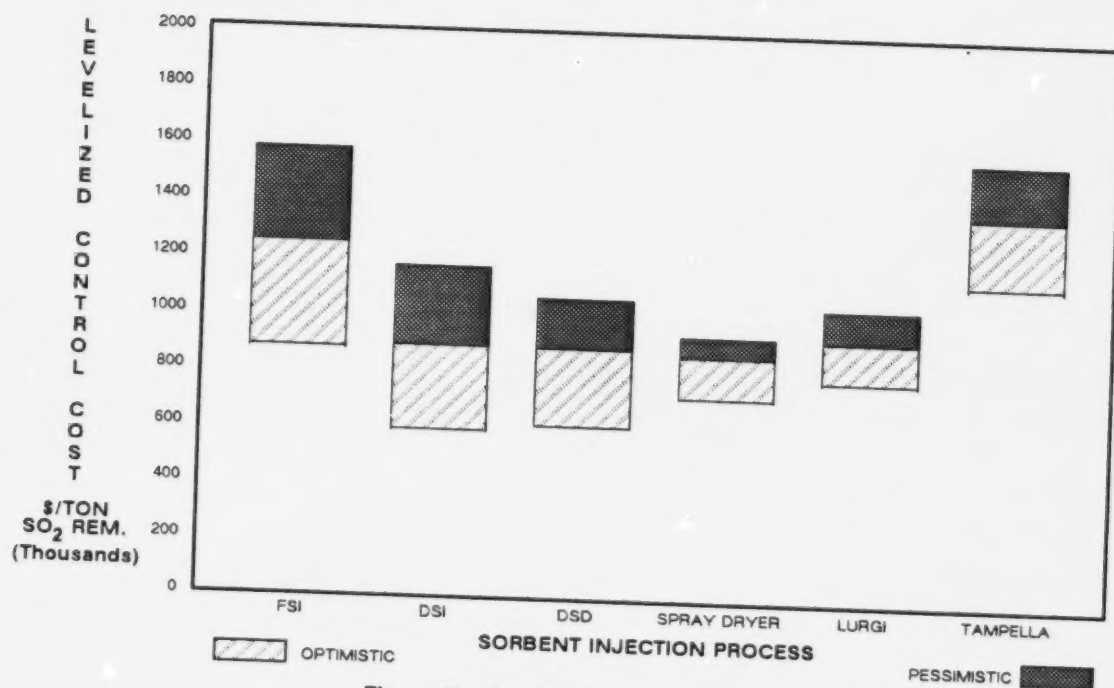


**Figure 7. Control Cost vs Design Assumptions**  
Virginia Power/Chesterfield 4

TABLE 1. DESIGN BASIS

General Plant Information

|                | <u>Net MW (Summer)</u> | <u>Net Plant<br/>Heat Rate (Btu/kWhr)</u> | <u>Calculated Duct<br/>Residence Time<br/>(Seconds)*</u> |
|----------------|------------------------|-------------------------------------------|----------------------------------------------------------|
| Chesterfield 3 | 100                    | 11,119                                    | 1.6                                                      |
| Chesterfield 4 | 166                    | 9,765                                     | 1.5                                                      |
| Chesterfield 5 | 326                    | 9,803                                     | 1.0                                                      |
| Chesterfield 6 | 658                    | 9,696                                     | nil                                                      |
| Possum Point 3 | 101                    | 10,727                                    | 1.5                                                      |
| Possum Point 4 | 221                    | 9,623                                     | 1.7                                                      |
| TOTAL          | 1,572                  |                                           |                                                          |

\*Ductwork from AH to ESP - at full load conditions.

Fuel Design Basis: Capital costs based on 1.5 wt. % S coal.  
Operating costs based on 1.0 wt. % S coal.

Fly Ash Disposal System Modifications: Replace the existing wet fly ash system with a new larger dry pneumatic spent sorbent/fly ash system for all six processes.

Sorbent Injection System Design

Carbonate Sorbent Pulverizing Location: On-site (Pulverized to 90% less than 325 mesh)  
Lime Sorbent Hydration Location: On-site (Hydrated product is typically 90% less than 200 mesh)  
Sorbent Bulk Storage Capacity: 10 days

Economic Basis

|                | <u>Average Plant<br/>Capacity Factor<br/>(%)</u> | <u>Levelized<br/>Fixed Charge<br/>Rate (%)</u> | <u>Remaining<br/>Plant Life*<br/>(Years)</u> |
|----------------|--------------------------------------------------|------------------------------------------------|----------------------------------------------|
| Chesterfield 3 | 62                                               | 24.0                                           | 10                                           |
| Chesterfield 4 | 79                                               | 16.2                                           | 18                                           |
| Chesterfield 5 | 70                                               | 14.7                                           | 22                                           |
| Chesterfield 6 | 62                                               | 13.8                                           | 27                                           |
| Possum Point 3 | 59                                               | 19.7                                           | 13                                           |
| Possum Point 4 | 62                                               | 15.3                                           | 20                                           |

Minimum Acceptable Return (MAR) = 10.0%.

Inflation Rate = 5.0%.

Delivered Sorbent Costs (\$/ton):

Limestone = \$16.00

Quicklime = \$55.00

Atmospheric Hydrated Lime = \$58.00

Marginal Power Cost = \$0.029/kWh (on-peak)

\* Based on unofficial retirement dates for each unit and for study purposes only.

TABLE 2. SO<sub>2</sub> CONTROL PROCESS DESIGN CHARACTERISTICS

| Process               | Purchased Sorbent | Design Sorbent Performance (1.5 wt% S Coal) |                             | Operating Sorbent Performance (1.0 wt% S Coal) |                             | Sorbent Injection Location       | Type of Absorber/Reactor Added |
|-----------------------|-------------------|---------------------------------------------|-----------------------------|------------------------------------------------|-----------------------------|----------------------------------|--------------------------------|
|                       |                   | Ca/S Molar Ratio                            | SO <sub>2</sub> Removal (%) | Ca/S Molar Ratio                               | SO <sub>2</sub> Removal (%) |                                  |                                |
| 1. FSI                | Quicklime         | 3.0                                         | 52                          | 2.0                                            | 40                          | Upper Furnace                    | None                           |
| 2. DSI                | Quicklime         | 2.0                                         | 52                          | 2.0                                            | 50*                         | Ductwork Between AH & ESP        | None                           |
| 3. DSD                | Quicklime         | 1.5                                         | 52                          | 1.5                                            | 50                          | Ductwork Between AH & ESP        | None                           |
| 4. Spray Dryer        | Quicklime         | 1.4                                         | 90                          | 1.4                                            | 90#                         | Absorber Vessel Between AH & ESP | Spray Dryer                    |
| 5. Lurgi              | Quicklime         | 1.5                                         | 95                          | 1.5                                            | 95                          | Absorber Vessel Between AH & ESP | Circulating Fluid Bed Absorber |
| 6. Tampella Limestone |                   | 1.7                                         | 80                          | 1.7                                            | 80                          | Upper Furnace                    | Activation Reactor             |

\*Best case based on limited pilot data.

#Based on vendor proposals with a 500 SCA ESP.



TABLE 3. PROCESS APPLICABILITY AND LEVEL OF DEVELOPMENT

| Process                      | Level of Development                              | Advantages                                                                                                                                                                                                                                                                                                        | Disadvantages                                                                                                                                                                                                                            |
|------------------------------|---------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 1. Furnace Sorbent Injection | Demo Scale - U.S. & Canada<br>Commercial - Europe | <ul style="list-style-type: none"> <li>At a larger scale of development than DSI or OSD</li> <li>Risk of corrosion lower due to partial humidification</li> <li>Requires less water</li> <li>Low Capital Cost - Good retrofit application</li> <li>Can use either a limestone or hydrated lime sorbent</li> </ul> | <ul style="list-style-type: none"> <li>Potential for convective pass fouling</li> <li>Lower sorbent utilization</li> <li>Capable of only 50% SO<sub>2</sub> removal</li> </ul>                                                           |
| 2. Duct Sorbent Injection    | Demo Scale planned - U.S. & Canada                | <ul style="list-style-type: none"> <li>Low Capital Cost - Good retrofit application</li> </ul>                                                                                                                                                                                                                    | <ul style="list-style-type: none"> <li>Requires humidification to within 20-30°F of dewpoint</li> <li>Potential for solids deposition on duct walls or internals</li> <li>Capable of only 50% SO<sub>2</sub> removal</li> </ul>          |
| 3. Duct Spray Drying         | Demo Scale - U.S.                                 | <ul style="list-style-type: none"> <li>Requires only partial humidification</li> <li>Low Capital Cost - Good retrofit application</li> </ul>                                                                                                                                                                      | <ul style="list-style-type: none"> <li>Requires slurry drying in a short residence time (1-2 seconds)</li> <li>Potential for solids deposition on duct walls or internals</li> <li>Capable of only 60% SO<sub>2</sub> removal</li> </ul> |
| 4. Lime Spray Drying         | Commercial - 18 units in US                       | <ul style="list-style-type: none"> <li>Capable of 90-95% SO<sub>2</sub> removal with fabric filter</li> <li>Applicable to new or retrofit units</li> <li>Well-proven commercial process on low sulfur coals</li> </ul>                                                                                            | <ul style="list-style-type: none"> <li>Requires large absorber vessel w/10-12 seconds residence time</li> <li>SO<sub>2</sub> removal with an ESP is less than that with a fabric filter</li> </ul>                                       |
| 5. Lurgi CFB Absorption      | Commercial - 4 units in West Germany              | <ul style="list-style-type: none"> <li>Capable of 90-95% SO<sub>2</sub> removal</li> <li>Applicable to new or retrofit units</li> </ul>                                                                                                                                                                           | <ul style="list-style-type: none"> <li>Requires large absorber vessel w/4-6 seconds residence time</li> <li>Requires a mechanical dust collector ahead of the ESP</li> </ul>                                                             |
| 6. Tampella LIFAC            | Semi-Commercial - 1 unit in Finland               | <ul style="list-style-type: none"> <li>Capable of 80% SO<sub>2</sub> removal - good application for a retrofit unit</li> <li>Uses a limestone sorbent</li> </ul>                                                                                                                                                  | <ul style="list-style-type: none"> <li>Requires large absorber vessel w/4-8 seconds residence time</li> <li>Requires recycle of 10-20% of flue gas for reheating</li> </ul>                                                              |

TABLE 4. CONTROL COST VERSUS DESIGN ASSUMPTIONS  
CHESTERFIELD UNIT 4

| PROCESS        | DESIGN CASE     | Design Performance  |                             | Operating Performance |                             | Humidification or Absorber/Reactor Capital Cost* (% of Base Capital Cost) | Type of Spent Sorbent/Fly Ash Disposal Method | Levelized Control Cost (\$/Ton SO <sub>2</sub> Removed) | % Change in Control Cost From Base |
|----------------|-----------------|---------------------|-----------------------------|-----------------------|-----------------------------|---------------------------------------------------------------------------|-----------------------------------------------|---------------------------------------------------------|------------------------------------|
|                |                 | Sorbent Performance | SO <sub>2</sub> Removal (%) | Sorbent Performance   | SO <sub>2</sub> Removal (%) |                                                                           |                                               |                                                         |                                    |
| 1. FSI         | Optimistic Base | Ca/S Molar Ratio    | 52                          | Ca/S Molar Ratio      | 40                          | 0.75                                                                      | Wet                                           | 836                                                     | - 32%                              |
|                | Pessimistic     | 3.0                 | 52                          | 2.0                   | 40                          | 1.00                                                                      | Dry                                           | 1,221                                                   | ---                                |
|                |                 | 4.0                 | 52                          | 3.0                   | 40                          | 1.25                                                                      | Dry                                           | 1,570                                                   | + 29%                              |
| 2. DSI         | Optimistic Base | 1.5                 | 52                          | 1.5                   | 50                          | 0.75                                                                      | Wet                                           | 576                                                     | - 36%                              |
|                | Pessimistic     | 2.0                 | 52                          | 2.0                   | 50                          | 1.00                                                                      | Dry                                           | 894                                                     | ---                                |
|                |                 | 3.0                 | 52                          | 3.0                   | 50                          | 1.25                                                                      | Dry                                           | 1,167                                                   | + 31%                              |
| 3. DSD         | Optimistic Base | 1.2                 | 52                          | 1.2                   | 50                          | 0.75                                                                      | Wet                                           | 606                                                     | - 31%                              |
|                | Pessimistic     | 1.5                 | 52                          | 1.5                   | 50                          | 1.00                                                                      | Dry                                           | 878                                                     | ---                                |
|                |                 | 2.0                 | 52                          | 2.0                   | 50                          | 1.25                                                                      | Dry                                           | 1,061                                                   | + 21%                              |
| 4. Spray Dryer | Optimistic Base | 1.2                 | 90                          | 1.2                   | 90                          | 0.75                                                                      | Wet                                           | 710                                                     | - 18%                              |
|                | Pessimistic     | 1.4                 | 90                          | 1.4                   | 90                          | 1.00                                                                      | Dry                                           | 861                                                     | ---                                |
|                |                 | 1.7                 | 90                          | 1.7                   | 90                          | 1.25                                                                      | Dry                                           | 937                                                     | + 9%                               |
| 5. Lurgi       | Optimistic Base | 1.3                 | 95                          | 1.3                   | 95                          | 0.75                                                                      | Wet                                           | 778                                                     | - 17%                              |
|                | Pessimistic     | 1.5                 | 95                          | 1.5                   | 95                          | 1.00                                                                      | Dry                                           | 935                                                     | ---                                |
|                |                 | 2.0                 | 95                          | 2.0                   | 95                          | 1.25                                                                      | Dry                                           | 1,040                                                   | + 11%                              |
| 6. Tampella    | Optimistic Base | 1.4                 | 80                          | 1.4                   | 80                          | 0.75                                                                      | Wet                                           | 1,142                                                   | - 18%                              |
|                | Pessimistic     | 1.7                 | 80                          | 1.7                   | 80                          | 1.00                                                                      | Dry                                           | 1,388                                                   | ---                                |
|                |                 | 2.5                 | 80                          | 2.5                   | 80                          | 1.25                                                                      | Dry                                           | 1,573                                                   | + 13%                              |

\* Humidification Capital Cost - FSI, DSI and DSD  
Absorber/Reactor Capital Cost - Spray Dryer, Lurgi, Tampella

LIME/LIMESTONE FGD AND SORBENT INJECTION TECHNOLOGY  
RETROFIT DIFFICULTY AT 60 COAL-FIRED UTILITY POWER PLANTS

by: Thomas E. Emmel  
Radian Corporation  
P.O. Box 13000  
Research Triangle Park, NC 27709

Norman Kaplan  
U.S. Environmental Protection Agency  
Air and Energy Engineering Research Laboratory  
Research Triangle Park, NC 27711

ABSTRACT

This paper presents the interim results from an ongoing program conducted under the National Acid Precipitation Assessment Program (NAPAP) by the U.S. Environmental Protection Agency's (EPA) Air and Energy Engineering Research Laboratory. The objective of this research program is to significantly improve engineering cost estimates for retrofitting  $\text{SO}_2$  and  $\text{NO}_x$  controls to existing coal-fired utility boilers. Under this program, both detailed and simplified procedures were developed and are being used to estimate the site-specific costs for retrofitting  $\text{SO}_2$  and  $\text{NO}_x$  controls at coal-fired utility boilers.

Detailed procedures have been used to estimate site-specific retrofit factors and costs for various  $\text{SO}_2/\text{NO}_x$  emission reduction options at the 12 plants for which site visits were conducted. Information developed from these site visits was used to refine the detailed procedures and to develop simplified retrofit estimating procedures. The simplified procedures are being used to estimate retrofit costs at the top 200  $\text{SO}_2$  emitting coal-fired power plants in the 31-state eastern region. The difficulty of retrofitting conventional lime/limestone (L/LS) FGD and sorbent injection technologies at 60 coal-fired power plants that have been evaluated is discussed.

## INTRODUCTION

Detailed and simplified procedures were developed and are being used to estimate the site-specific costs for retrofitting SO<sub>2</sub> and NO<sub>x</sub> controls at coal-fired utility boilers. The detailed procedures have been used to estimate site-specific retrofit factors and costs for various SO<sub>2</sub>/NO<sub>x</sub> emission reduction options at 12 plants for which site visits were conducted (1).<sup>x</sup> Preliminary results of this effort were presented at the 1986 meeting (Tenth Symposium on Flue Gas Desulfurization). Information developed from these site visits was used to refine the detailed procedures and to develop simplified retrofit estimating procedures.

The simplified procedures have been used to estimate the retrofit difficulty of L/LS FGD, duct spray drying (DSD), and furnace sorbent injection (FSI) at 48 coal-fired utility plants. Six of these plants were visited and the simplified procedure results were compared to the detailed procedure results and cost/performance estimates provided by the utility companies. Based on this review, the simplified procedures were modified and the cost/performance estimates for the 48 plants have been finalized. Cost/performance estimates for the remaining top 200 plants are now being developed using the simplified procedures.

## METHODOLOGY

The Integrated Air Pollution Control System (IAPCS) cost model (2) was used to develop the control technology cost estimates. However, the model cost algorithms do not reflect the effect of a retrofit on control cost and performance. To adjust the IAPCS model algorithms for retrofits, retrofit factors and cost adders were developed for each of the control technologies, adjusting the cost estimates to reflect specific situations. The Electric Power Research Institute (EPRI) "Retrofit FGD Cost Estimating Guidelines" report (3) was used to develop retrofit factors and scope adders for L/LS FGD. To the extent possible, the information and methodology contained in the EPRI Guidelines were used for the sorbent injection technologies.

The general approach used to adjust the IAPCS cost and performance estimates was to develop retrofit difficulty multipliers (retrofit factors) and scope adder costs. Retrofit factors adjust model process area costs to reflect the effects of:

- o site access and congestion,
- o underground obstructions, and
- o distances between process areas.

Scope adders adjust the cost model estimates by adding costs that were not addressed in the model algorithms. Typical scope adders that are not included in cost algorithms developed for new plant control systems or in retrofit control system bases include:

- o chimney liner or new chimney,
- o demolition/relocation of existing facilities,
- o additional particulate control facilities, and
- o conversion of wet ash handling systems to dry handling.



The retrofit factors and scope adders were then input to IAPCS to adjust the cost model results to reflect the effects of a site-specific retrofit situation.

For L/LS FGD, the retrofit factors which most significantly affect control costs are the SO<sub>2</sub> absorber process area access and congestion and the flue gas duct length to the SO<sub>2</sub> absorber area. For the sorbent injection technologies, particulate control type, size, and configuration are the most significant factors. Therefore, it was necessary to obtain aerial photographs and/or plant plot plans and Energy Information Agency (EIA) Form 767 data for each plant. The plant photograph/plot plan was used to locate equipment and identify space limitations. The EIA Form 767 data provided coal, boiler, and particulate control data and confirmed existing equipment configurations.

To develop the retrofit factors for L/LS FGD for each boiler evaluated, the following steps were taken:

- o identify location of SO<sub>2</sub> absorber, sorbent preparation/storage, and waste handling areas;
- o estimate flue gas ducting length;
- o estimate SO<sub>2</sub> absorber area and flue gas ducting access/congestion factors;
- o identify need for other scope adders (new chimney and wet-to-dry fly ash handling conversion); and
- o select general facilities percentage based on existing equipment demolition and replacement.

To develop the retrofit difficulty factor for FSI and DSD for each boiler evaluated, the following steps were taken:

- o identify location of sorbent storage/preparation and waste handling areas;
- o evaluate reusability of current particulate controls;
- o estimate access/congestion factor for existing/new particulate controls and sorbent preparation area; and
- o identify need for other scope adders (wet-to-dry fly ash handling system conversion and flue gas ducting demolition/addition).

Table 1 presents the percent increase of base system costs due to these retrofit factors and scope adders. For L/LS FGD, the retrofit difficulty factor can range from 1.15 (15% increase) for the easiest retrofit to greater than 2.0 for the most difficult retrofit. For FSI and DSD, the retrofit difficulty factor can range from 1.0 for the easiest retrofit to greater than 2.0 for difficult retrofit situations requiring new particulate controls and extensive flue gas ducting.

Table 2 presents an example of the plant operational data used to estimate L/LS FGD and FSI/DSD retrofit factors. This information was obtained from EIA Form 767. Figure 1 presents an example plant plot plan developed from an aerial photograph.

## L/LS FGD RETROFIT DIFFICULTY RESULTS

Before presenting the L/LS FGD retrofit difficulty and cost results for the 60 plants evaluated to date, a brief example is presented to describe the procedures used. These procedures were applied to each of the boilers at the 60 plants.

### EXAMPLE ANALYSIS

Based on the plant information presented in Table 2 and the plot plan presented in Figure 1, an FGD retrofit difficulty factor was developed for the southernmost boiler. Table 3 presents the retrofit factor data sheet for this 450 MW boiler. Space is available adjacent to the retrofit ESPs (south of the unit) to locate the sorbent handling, flue gas handling, SO<sub>2</sub> scrubbing areas, and a new chimney. The waste handling area would be located farther south and east on an old ash disposal site. Figure 1 shows the location of the FGD process areas. A short run (0 - 100 ft or 0 - 30 m) of flue gas ducting would be required to tie the SO<sub>2</sub> absorbers into the existing flue gas ducting coming from the ESPs. A moderate access/congestion factor was selected for the SO<sub>2</sub> absorber area due to the restricted access caused by the combustor turbines, existing ESPs, and the river. A low site access/congestion factor was assigned to the flue gas ducting area because of the short duct runs. A low general facilities factor was assigned because no demolition/relocation of existing facilities was required. It was assumed that a new chimney would be required, because access to the existing one is difficult and this chimney is shared with another unit. Also, it was assumed that the existing wet fly ash handling system could be converted to a dry system so that the fly ash could be used to stabilize the FGD waste.

These retrofit parameter inputs result in a moderate overall retrofit difficulty factor of 1.34. This factor is input to the IAPCS cost model with the general facilities percentage to obtain a site specific retrofit cost estimate. For this example case, a capital cost estimate of \$75 million (April 1987 \$) or \$189/kW was obtained. This estimate compares well with the estimated cost of \$113 million based on a much more detailed cost estimate for this unit provided by the utility company. The \$113 million estimate is based on firing a 5.5% sulfur coal versus the study basis of a 2% sulfur coal.

### SIXTY PLANT RESULTS

The above example summarizes the methodology used to evaluate all coal-fired boilers at the 60 plants evaluated to date. Table 4 summarizes the L/LS FGD retrofit difficulty factors developed for 57 of the 60 plants. Of the 191 boilers evaluated, 35% had a low retrofit difficulty, 55% had a moderate retrofit difficulty, and 10% had a high retrofit difficulty. The overall retrofit difficulty factors ranged from 1.19 to over 2.

Although retrofit difficulty directly affects the capital cost of control, many other factors will influence the selection of the least cost control method. Of particular importance is the level of control, the unit size and capacity factor, and the coal characteristics (sulfur content and price relative to alternative coals). Table 5 summarizes the SO<sub>2</sub> cost effectiveness (April 1987 \$) for the 191 boilers for which L/LS FGD retrofit factors and costs were developed.

Cost effectiveness<sup>2</sup> as used in this paper means: levelized annual cost per ton<sup>1</sup> of pollutant removed.<sup>2</sup> Therefore, a high cost effectiveness value indicates poor process economics, and a low cost effectiveness value indicates good process economics. Only 17% of the boilers are very cost-effective to scrub (<\$1000/ton<sup>1</sup>); 43% are moderately cost-effective to scrub; and 40% are not cost effective to scrub (>\$2000/ton).

The cost effectiveness results were analyzed with respect to the effect of retrofit difficulty, coal sulfur, and boiler capacity factor and size. Based on this analysis, surprisingly, it was found that very few of the boilers that had high retrofit<sup>2</sup> difficulty also had very high cost per ton of SO<sub>2</sub> removed (cost effectiveness<sup>2</sup>). Of the 75 boilers (75 of 191 boilers) which have high cost effectiveness values, over 50% are small and already burn low sulfur coals (<2% sulfur). Approximately 20% of the boilers evaluated to date had low capacity factors (<40%).

The cost effectiveness results presented here are biased on the high side for two reasons. First, it was assumed that the boiler would continue to fire the same coal after retrofit of FGD. This is not likely to happen. Instead a lower cost higher sulfur coal would be fired or compliance would be met using another control technology (sorbent injection or coal switching). If a higher sulfur coal is fired, the cost effectiveness values for the boilers would drop significantly. If it is assumed that boilers burning low sulfur coals would switch to higher sulfur coals resulting in a 50% reduction in the cost effectiveness value, the number of boilers having high cost effectiveness drops from 40 to 27% of the 191 boilers evaluated. As shown in Table 6, 8 boilers shift from moderate to low cost-effectiveness values and 24 boilers shift from high to moderate cost-effectiveness values (April 1987 \$).

The second factor biasing the cost effectiveness results presented in Table 5 is that the costs are based on a stand-alone FGD system at each boiler, except where the flue gases from several boilers have already been combined before the chimney (43 boilers). As part of the study, a low cost FGD system was estimated for all of the plants. This system assumed that, where feasible, boilers at the plant would be ducted together into a single FGD system. Applying this option at 22 plants having small boilers (<250 MW) shifts 22 boilers to having a cost effectiveness less than \$2000/ton. Also, 27 boilers that have moderate cost effectiveness values shift to having cost effectiveness of less than \$1000/ton. As shown in Table 6, this results in reducing the number of boilers with high cost effectiveness values from 40 to 28% of the 191 boilers evaluated.

#### SORBENT INJECTION RETROFIT DIFFICULTY RESULTS

A number of sorbent injection technologies are being developed as low capital cost SO<sub>2</sub> control processes. Only calcium-based sorbents combined with flue gas humidification were considered in this study. The specific processes considered were furnace sorbent injection (FSI) with humidification and duct spray drying (DSD). Two configurations of duct spray drying were considered: sorbent injection in the duct and sorbent injection in the first ESP section (E-SOX).

The FSI process uses dry injection of a calcitic hydrate material followed by in-duct humidification prior to the particulate control device. The U.S. EPA is demonstrating this process at the Ohio Edison Edgewater plant and the Richmond

<sup>1</sup> 1 ton = 0.9072 metric ton

<sup>2</sup> Capital recovery factor of 0.17 and operating/maintenance levelization factor of 1.75.



Power and Light Whitewater Valley station. The DSD and E-SOX processes inject a lime slurry into the hot flue gas prior to or in the first ESP section. The duct injection process is actively being developed by several companies and research organizations. The E-SOX process is based on the successful development of advanced ESP technology that would allow for a typical three-field ESP to be upgraded so that only the final two fields would be required for particulate removal. This process is being demonstrated on a 5 MW pilot scale facility by EPA and the Ohio Coal Development Office at the Ohio Edison Burger plant.

The major factors affecting the retrofit of these technologies are the size and type of existing particulate control device, flue gas residence time, and duct configuration after the air heater and before the particulate control device. Sufficient space and residence time are required for humidification in the case of FSI or lime slurry droplet drying for DSD. If the particulate control device is an ESP, it must be large and efficient enough to be easily upgraded for adequate particulate control or be easy to enlarge. Under this study, one of the above technologies was considered applicable if:

1. the ESP had a specific collection area (SCA<sup>1</sup>) greater than 250 ft<sup>2</sup>/1000 acfm of flue gas,
2. the existing particulate control device was a fabric filter, or
3. the ESP had an SCA less than 250 but sufficient space was available to add plate area.

For a number of situations, the cost of adding a new fabric filter was evaluated.

Table 7 presents the retrofit factor data developed for the example unit characterized in Table 2 and Figure 1. The sorbent handling area was assumed to be located in the open area south of the ESPs. Because this area is open, a low access/congestion retrofit difficulty was assigned to the sorbent handling area and to the ESP upgrade. Because the ESP has a large SCA of 646, no major ESP upgrades would be necessary except those specific to the technology (e.g., humidification with FSI). Additionally, because this unit has a wet ash handling system, conversion from a wet to dry system is necessary to prevent plugging of the sluice lines. No significant additional flue gas ducting is required because no additional plate area or new particulate controls are needed. Only a small amount of existing duct work is assumed to be removed to make necessary water/sorbent injection modifications. The resulting inputs to the IAPCS cost model are the ESP retrofit difficulty factor of 1.13 and a scope adder capital cost of \$3.7 million.

However, at this same plant, the other boilers which range in size from 100 to 225 MW have small ESPs (SCA of 205-247). The ESPs were retrofitted in the 1970s and require SO<sub>2</sub> conditioning to meet emission standards. These ESPs were built above the turbine houses due to space constraints. Flue gas residence time before the ESPs is short and plate area addition would be difficult. As a result, use of the sorbent injection technologies may not be feasible at a reasonable cost.

This example summarizes the methodology used to develop particulate control and reagent preparation access/congestion factors and scope adders that were input to the IAPCS cost model. Table 8 presents the particulate control characteristics

<sup>1</sup>SCA unit is in ft<sup>2</sup>/1000 acfm -- 1 ft<sup>2</sup>/1000 acfm = 0.1967 sec/m



for the 60 plants evaluated to date. Of the 191 boilers evaluated, 18% have very small ESPs (<200 SCA), 38% have marginal size ESPs (200-300 SCA), and 44% have large ESPs (>300 SCA) and fabric filters.

All of the boilers having very small ESPs are unlikely to have significant flue gas residence time between the air heater and ESP. Therefore, the addition of ESP plate area or the retrofit of a new fabric filter would be required. Obviously, these costs significantly increase the capital requirements of the sorbent injection technologies. However, the same type of retrofit costs would be required to switch these units to a lower sulfur coal. For those boilers having marginal size ESPs, only about 25% have sufficient flue gas residence time between the air heater and ESP to allow for lime slurry droplet drying without major duct configuration modifications. Therefore, most of these boilers would be better candidates for application of FSI and E-SOX than for DSD. For the boilers having ESPs larger than 300 SCA or fabric filters, almost all have moderate to long flue gas residence times between the air heater and the ESP. These boilers would be good candidates for application of any of the sorbent injection technologies.

The potential for retrofit of the sorbent injection technologies is good, particularly where ESPs having an SCA larger than 250 can be easily upgraded to accommodate the increased particulate loading. Of the 191 boilers, 84 boilers are estimated to have moderate to high L/LS FGD cost-effectiveness values (>\$1500/ton). These boilers are good potential candidates for applying the sorbent injection technologies for the following reasons.

1. Of these boilers, 87% (73 boilers) are less than 250 MW in size.
2. Of these boilers, 68% (57 boilers) fire coal with a sulfur content less than 2%.
3. Of these boilers, 50% (42 boilers) have a capacity factor less than 40%.
4. Of these boilers, 50% (42 boilers) have ESPs with SCAs greater than 250 or have fabric filters. The majority of the other boilers are cyclone boilers firing high sulfur coals.

#### CONCLUSIONS

The results of the 60 plants evaluated indicate that FGD retrofit capital costs will be 20 to 100% higher than for new plant applications. The incremental cost of retrofit difficulty can be significant. For a small boiler (100 MW) with an average capacity factor of 50%, an average retrofit difficulty factor of 1.5 increases the cost of power generation (\$/MW-hr) by 31% over a new plant FGD system (retrofit difficulty of 1.0). For most small boilers there will be the opportunity to install a large, combined FGD system to reduce costs through economies of scale. However, unit capacity factor has a much greater impact on the cost of power relative to retrofit difficulty. For most small boilers, the unit capacity factors are low because of the higher than average unit heat rate. The addition of an FGD system will further degrade the unit heat rate which will further reduce the unit dispatch order relative to large units.

The potential for retrofit of the sorbent injection technologies is good where ESP SCA is greater than 250 and the boiler is small, has a moderate to low capacity factor, and fires a 1-2% sulfur coal. Although switching to a low sulfur coal (<1% S) is certainly an option, this will generally require extensive ESP upgrade, and the price of low sulfur coal may increase significantly if large reductions of SO<sub>2</sub> are mandated. Because the calcium based sorbent injection

technologies typically include flue gas humidification, it appears that significant ESP upgrade costs will not be incurred for most boilers applying these technologies.

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This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.

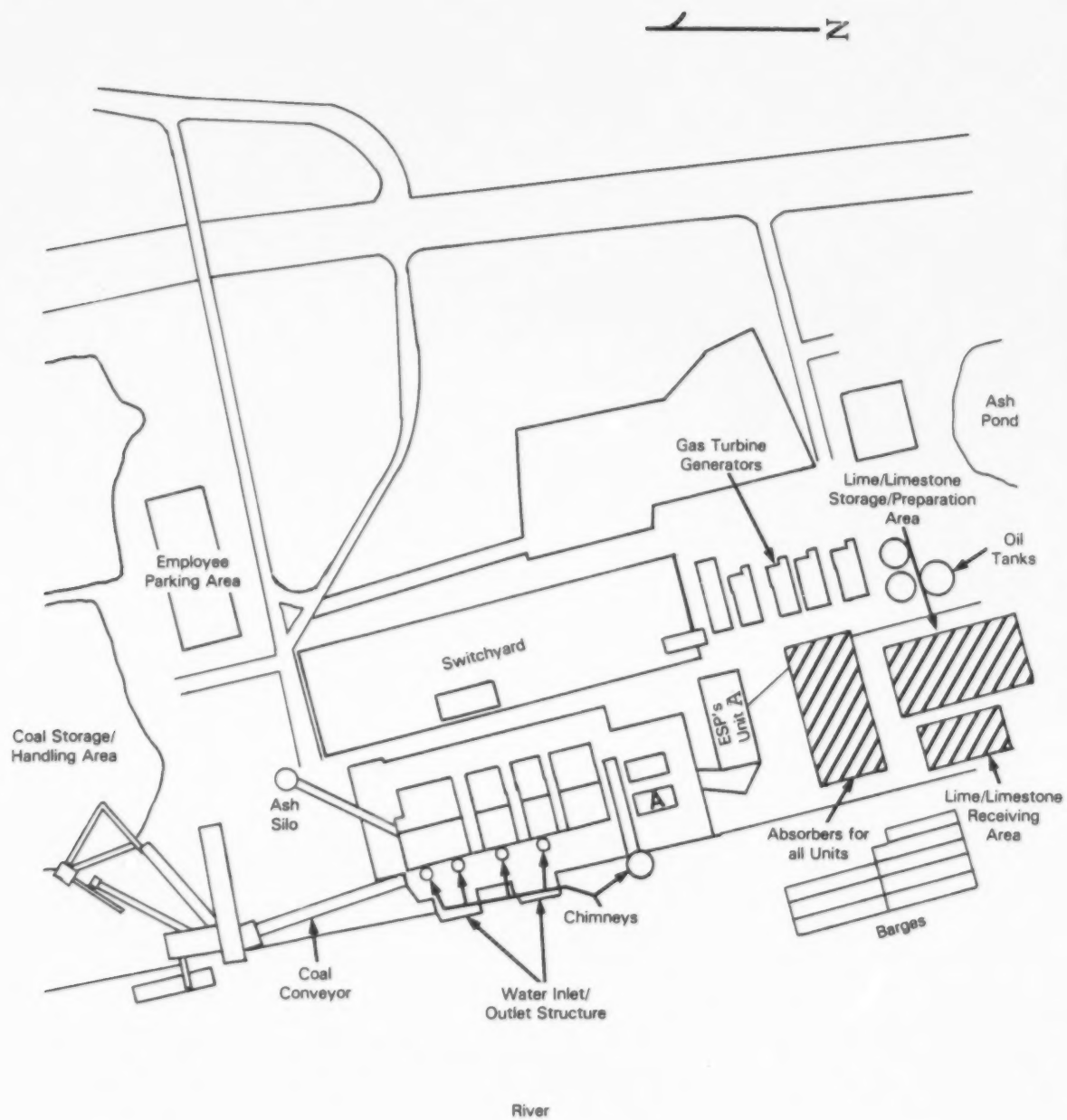


Figure 1. Example Plant A



TABLE 1. EFFECT OF RETROFIT FACTORS ON L/LS FGD AND SORBENT INJECTION TECHNOLOGY CAPITAL COSTS

| Cost Multipliers and Scope Adders | Approximate Increase in Base Cost (%) |         |
|-----------------------------------|---------------------------------------|---------|
|                                   | L/LS FGD                              | FSI/DSD |
| Site Access and Congestion        |                                       |         |
| Low                               | 8                                     | 0       |
| Medium                            | 25                                    | 15      |
| High                              | 42                                    | 25      |
| New Flue Gas Ducting              |                                       |         |
| <100 ft*                          | 2                                     | NA      |
| 100-300 ft                        | 6                                     | NA      |
| 300-600 ft                        | 12                                    | NA      |
| 600-1000 ft                       | 18                                    | NA      |
| 1000 + ft                         | 35                                    | NA      |
| Wet-to-Dry Fly Ash Handling       | 7                                     | 30-60   |
| New Chimney                       | 7                                     | NA      |
| General Facilities                |                                       |         |
| Low                               | 5                                     | NA      |
| Medium                            | 10                                    | NA      |
| High                              | 15                                    | NA      |
| ESP Upgrade                       | NA                                    | 30-60   |
| ESP Upgrade + Plate Area          | NA                                    | >100    |

\* 1 ft = 0.3048 m

TABLE 2. UNIT OPERATING DATA

|                                                | Example Unit |
|------------------------------------------------|--------------|
| Boiler Number                                  | A            |
| Generating Capacity (MW-each)                  | 450          |
| Installation Date                              | 1969         |
| Firing Type                                    | Tangential   |
| Coal Sulfur Content (%)                        | 2.2          |
| Coal Heating Value (Btu/lb)*                   | 11000        |
| Coal Ash Content (%)                           | 16.0         |
| Fly Ash System                                 | Wet Sluice   |
| Ash Disposal Method                            | On-Site      |
| Stack Number                                   | Shared       |
| Coal Delivery Methods                          | Barge        |
| <u>Particulate Matter</u>                      |              |
| Type                                           | ESP          |
| Installation Date                              | 1969/79      |
| Emission (lb/10 <sup>6</sup> Btu) <sup>+</sup> | 0.03         |
| Removal Efficiency                             | 99.5         |
| Design Specification                           |              |
| Sulfur (%)                                     | 2.5          |
| SCA (ft <sup>2</sup> /1000 acfm) <sup>#</sup>  | 646          |
| Outlet Temperature (°F) <sup>§</sup>           | 300          |

\* 1 Btu/lb = 0.2339 J/g

+ 1 lb/10<sup>6</sup> Btu = 4.186 x 10<sup>-6</sup> g/J

# 1 ft<sup>2</sup>/1000 acfm = 0.1967 sec/m

§ °C = 5/9 (°F - 32)

TABLE 3. L/LS FGD RETROFIT FACTOR DATA SHEET

| Retrofit Parameters                                          | Example Units |
|--------------------------------------------------------------|---------------|
| <u>Site Access/Congestion</u>                                |               |
| SO <sub>2</sub> Removal                                      | Medium        |
| Flue Gas Handling                                            | Low           |
| Duct Work Distance (ft)*                                     | 0-100         |
| <u>Scope Adjustments</u>                                     |               |
| Wet-to-Dry Ash Handling                                      | Yes           |
| New Chimney                                                  | Yes           |
| Other                                                        | No            |
| <u>General Facilities (%)</u>                                | 5             |
| <u>Retrofit Factor (calculated by simplified procedures)</u> |               |
| FGD System                                                   | 1.34          |

\* 1 ft = 0.3048 m

TABLE 4. L/LS FGD RETROFIT DIFFICULTY DISTRIBUTION

| State          | Number of Boilers | Boiler Population Retrofit Difficulty |                         |               |
|----------------|-------------------|---------------------------------------|-------------------------|---------------|
|                |                   | Low<br>1.19 - 1.33                    | Moderate<br>1.34 - 1.67 | High<br>>1.67 |
| Ohio           | 52                | 15                                    | 23                      | 14            |
| Missouri       | 21                | 7                                     | 14                      | 0             |
| Illinois       | 24                | 5                                     | 19                      | 0             |
| Pennsylvania   | 28                | 21                                    | 3                       | 4             |
| Kentucky       | 13                | 7                                     | 5                       | 1             |
| TVA (TN & AL)* | <u>53</u>         | <u>11</u>                             | <u>42</u>               | <u>0</u>      |
| Totals:        | 191               | 66                                    | 106                     | 19            |
| % of Total     |                   | 35                                    | 55                      | 10            |

\* Tennessee Valley Authority plants in Tennessee and Alabama.

TABLE 5. SUMMARY OF L/LS FGD COST EFFECTIVENESS RESULTS

| State          | Number of Boilers | SO <sub>2</sub> Cost-Effectiveness <sup>#</sup> |                         |                     |
|----------------|-------------------|-------------------------------------------------|-------------------------|---------------------|
|                |                   | Low<br><\$1000/ton <sup>+</sup>                 | Medium<br>1000-2000/ton | High<br>>\$2000/ton |
| Ohio           | 52                | 8                                               | 25                      | 19                  |
| Missouri       | 21                | 9                                               | 5                       | 7                   |
| Illinois       | 24                | 7                                               | 2                       | 15                  |
| Pennsylvania   | 28                | 2                                               | 22                      | 4                   |
| Kentucky       | 13                | 2                                               | 7                       | 4                   |
| TVA (TN & AL)* | <u>53</u>         | <u>5</u>                                        | <u>22</u>               | <u>26</u>           |
| Totals:        | 191               | 33                                              | 83                      | 75                  |
| % of Total     |                   | 17                                              | 43                      | 40                  |

<sup>#</sup> Levelized annual cost per ton of pollutant. Capital cost based on April 1987 \$. Capital recovery factor of 0.17 and operating/maintenance levelization factor of 1.75.

<sup>+</sup> 1 ton = 0.9072 metric ton.

\* Tennessee Valley Authority plants in Tennessee and Alabama.



TABLE 6. EFFECT OF SWITCHING TO HIGH SULFUR COAL AND COMBINED FGD SYSTEMS  
ON L/LS FGD COST-EFFECTIVENESS

| Scenario Description                                                          | Boilers<br>% of Total | Boiler SO <sub>2</sub> Cost-Effectiveness <sup>‡</sup> |             |
|-------------------------------------------------------------------------------|-----------------------|--------------------------------------------------------|-------------|
|                                                                               |                       | <\$1000/ton*                                           | >\$2000/ton |
| Individual FGD systems on each boiler and no switching to higher sulfur coals | Boilers<br>% of Total | 33                                                     | 83          |
| Individual FGD systems on each boiler and switching to higher sulfur coals    | Boilers<br>% of Total | 17                                                     | 43          |
| Combined FGD systems on small boilers and no switching to higher sulfur coals | Boilers<br>% of Total | 41                                                     | 99          |
|                                                                               |                       | 21                                                     | 52          |
|                                                                               |                       | 74                                                     | 64          |
|                                                                               |                       | 39                                                     | 33          |
|                                                                               |                       |                                                        | 28          |

\* Levilized annual cost per ton of pollutant. Capital cost based on April 1987 \$. Capital recovery factor of 0.17 and operating/maintenance levelization factor of 1.75.

\* 1 ton = 0.9072 metric ton.

+ Assumes 50% reduction in cost-effectiveness for 48 boilers currently burning <2% sulfur coal due to shifting to high sulfur coal after retrofit of L/LS FGD system.

# Assumes that a combined FGD system was used at 49 boilers resulting in a significant decrease in cost-effectiveness values due to economies of scale of a large (>400 MW) FGD system versus a small system (<250 MW).

TABLE 7. DUCT SPRAY DRYING AND FURNACE SORBENT INJECTION  
RETROFIT FACTOR DATA

|                                                    | Example Unit |
|----------------------------------------------------|--------------|
| <u>Site Access/Congestion</u>                      |              |
| Reagent Preparation                                | Low          |
| ESP Upgrade                                        | Low          |
| New Fabric Filter                                  | NA           |
| <u>Scope Adders</u>                                |              |
| Change ESP Ash Discharge System<br>to Dry Handling | Yes          |
| Estimated Cost (1000\$)                            | 3,603        |
| Additional Duct Work (ft)                          | 0            |
| New Fabric Filter                                  | NA           |
| Estimated Cost (1000\$)                            | NA           |
| ESP Reuse Upgrade                                  | 0            |
| Estimated Cost (1000\$)                            | 0            |
| Duct Demolition Length (ft)                        | 50           |
| Demolition Cost (1000\$)                           | 92           |
| Total Cost (1000\$)                                | 3695         |
| <u>Retrofit Factors</u>                            |              |
| Reagent Preparation                                | 1.0          |
| ESP Upgrade                                        | 1.13         |
| New Fabric Filter                                  | NA           |

NA = not applicable

\* 1 ft = 0.3048 m

TABLE 8. PARTICULATE CONTROL DISTRIBUTION

| State                      | Number of Boilers | Boiler Population Particulate Control Characteristics |             |             |          |
|----------------------------|-------------------|-------------------------------------------------------|-------------|-------------|----------|
|                            |                   | <200 SCA*                                             | 200-300 SCA | 301-400 SCA | >400 SCA |
| Ohio                       | 52                | 7                                                     | 19          | 9           | 13       |
| Missouri                   | 21                | 1                                                     | 8           | 9           | 3        |
| Illinois                   | 24                | 14                                                    | 8           | 0           | 2        |
| Pennsylvania               | 28                | 4                                                     | 18          | 0           | 3        |
| Kentucky                   | 13                | 5                                                     | 6           | 2           | 0        |
| TVA (TN & AL) <sup>+</sup> | 53                | 4                                                     | 13          | 7           | 19       |
| Totals                     | 191               | 35                                                    | 72          | 27          | 40       |
|                            |                   |                                                       |             |             | 17       |

\* SCA - Specific collection area of electrostatic precipitator (square feet of plate area per 1000 actual cubic feet a minute of flue gas)

<sup>+</sup> Tennessee Valley Authority plants in Tennessee and Alabama.

### SESSION 3: SPRAY DRYER TECHNOLOGY

Chairman, Robert L. Glover, EPRI

#### RESULTS FROM THE TVA 10-MW SPRAY DRYER/ESP EVALUATION

CHARLES A. BROWN and GARY M. BLYTHE  
Radian Corporation  
Austin, Texas 78720

LYNN R. HUMPHRIES, RUSSELL F. ROBARDS, and RICHARD A. RUNYAN  
Tennessee Valley Authority  
Chattanooga, Tennessee 37402

RICHARD G. RHUDY  
Electric Power Research Institute  
Palo Alto, California 94303

#### ABSTRACT

To evaluate the feasibility of retrofitting spray dryers for flue gas desulfurization (FGD) at existing power plants, TVA has constructed and is operating a 10-MW spray dryer/ESP pilot plant to treat flue gas from a boiler firing medium- and high-sulfur coals. EPRI and Ontario Hydro are cofunding the project. Testing is scheduled to continue through 1989. The objectives of this test program are to: 1) measure  $\text{SO}_2$  removal in the spray dryer and ESP as a function of major operating variables, 2) evaluate the effects on an existing ESP of retrofitting a spray dryer FGD system upstream, and 3) evaluate the operability of a high-sulfur spray dryer FGD system.

This paper summarizes the results of the first one-and-a-half years of pilot scale tests. The results indicate that for a coal containing 4% sulfur and 0.05% chloride the technology can achieve  $\text{SO}_2$  removal efficiencies of 70% and 85% for reagent ratios (Ca/S) of 1.0 and 1.6, respectively. For a coal containing 4% sulfur and 0.25% chloride, overall  $\text{SO}_2$  removal increased to 80% and 92% at the same reagent ratios. For a medium-sulfur coal containing 2.5% sulfur and 0.25% chloride, overall  $\text{SO}_2$  removal levels of 82% and 99% were obtained at these reagent ratios, and 90%  $\text{SO}_2$  removal was achieved at a reagent ratio of 1.3. These performance data are all for operation of the spray dryer at an 18°F approach to adiabatic saturation and a flue gas inlet temperature of 320°F.

Particulate mass loading data indicate that emissions at the ESP outlet increase when the spray dryer is in service, rather than decrease as would be expected because of the resulting improved electrical conditions. The cause of the increased emissions appears to be re-entrainment, mostly from scouring of low-resistivity particles, and partially from plate rapping. With a flue gas velocity through the ESP of about 3.5 ft/sec, outlet emissions were measured at  $0.05 \text{ lb}/10^6 \text{ Btu}$  with 4 fields in service and a specific collection area (SCA) of  $412 \text{ ft}^2/\text{kacfm}$ . With 3 fields and an SCA of 311, outlet emissions were measured at  $0.10 \text{ lb}/10^6 \text{ Btu}$ , and with 2 fields and an SCA of 212, emissions were measured at  $0.66 \text{ lb}/10^6 \text{ Btu}$ .



## INTRODUCTION

The utility industry has a continuing interest in developing low cost alternatives to wet limestone scrubbing for flue gas desulfurization on coal-fired boilers in medium- and high-sulfur-coal applications. To this end, the Tennessee Valley Authority (TVA) conducted an evaluation of spray dryer technology for high-sulfur-coal applications at the 1-MW scale beginning in 1983. Both spray dryer/fabric filter and spray dryer/electrostatic precipitator (ESP) combinations were evaluated (1, 2). The results from those efforts indicated that the spray dryer/fabric filter combination could exceed 90%  $\text{SO}_2$  removal, and that an ESP was a viable alternate to a fabric filter as the particulate collection device in a spray dryer FGD system. The latter is particularly important for potential FGD retrofit situations where it would be desirable to use an existing ESP in a spray dryer FGD system rather than to install a new fabric filter.

To further develop spray dryer/ESP technology for high-sulfur-coal applications, TVA installed and is presently operating a 10-MW pilot spray dryer flue gas desulfurization system (FGD) that uses an ESP as the particulate collection device. The facility is co-sponsored by the Electric Power Research Institute (EPRI) and Ontario Hydro. Technical assistance also has been provided by the Kentucky Energy Cabinet Laboratory. The pilot facility is located at TVA's Shawnee Power Plant near Paducah, Kentucky. The test program began in March 1987, and the high-sulfur coal tests were completed in January 1988. Medium-sulfur coal testing began at that time.

The purpose of this program is to evaluate the feasibility of retrofitting spray dryer systems on plants burning medium- or high-sulfur coal. A primary objective of the pilot study has been to determine the effect of major operating variables on  $\text{SO}_2$  removal performance in both the spray dryer and the ESP. An equally important objective is to determine the effects of retrofitting a spray dryer FGD system on an existing ESP.

This paper includes a description of the pilot unit process and equipment,  $\text{SO}_2$  removal results, particulate removal results, and a discussion of operating experiences for the 17-month period from March 1987 through July 1988.

## BACKGROUND

In the U.S., there are 15 utility spray dryer FGD systems installed on over 5000 MW of generating capacity. All of these units are installed on boilers that fire coal with a sulfur content of less than 1.5%. While spray dryers have been successfully demonstrated for low-sulfur coal, this information is not directly applicable to medium- and high-sulfur coal applications. A key difference is that as coal sulfur increases, more  $\text{SO}_2$  must be transferred from the flue gas to the slurry droplets and reacted with lime. Furthermore, as the coal sulfur levels increase, higher percent  $\text{SO}_2$  removal levels may be required. Because of these effects, the lime requirement will be greater for medium- to high-sulfur coal applications, resulting in a reduction in the amount of product solids that can be recycled.

All of these effects make medium- and high-sulfur coals a more demanding application of spray dryer FGD technology. Operation at a lower approach temperature (i.e., closer approach to adiabatic saturation) may be required to achieve sufficient levels of  $\text{SO}_2$  removal. Few of the 15 existing utility spray dryer FGD systems are designed to operate at an approach temperature as low as 20°F, which may be necessary in high-sulfur coal applications. Other differences between the

the two applications are that ash from medium- and high-sulfur coals may contain less alkalinity, and hence will not likely contribute to  $\text{SO}_2$  removal by the system, and the flue gas may contain higher chloride levels. The increased chloride levels may have beneficial effects on system  $\text{SO}_2$  removal performance, but may adversely affect droplet drying and waste transport.

Of the 15 existing utility systems, only 3 use an ESP for particulate control. Consequently, there is relatively little information about the effects of a spray dryer FGD system on an ESP, and most of that information is only for low-sulfur-coal applications. Thus, there was a need to demonstrate spray dryer FGD technology in a system with an ESP where medium- to high-sulfur-coal was fired.

### PROCESS DESCRIPTION

A simplified process flow diagram for the 10-MW pilot unit is illustrated in Figure 1. A slipstream of about 35,000 acfm is withdrawn from the Shawnee Unit #9 boiler outlet duct, just downstream of mechanical collectors. A venturi measures flue gas flow at the pilot system inlet to allow precise control of the gas rate. A heat exchanger controls the inlet flue gas temperature. Inlet  $\text{SO}_2$  concentration is determined by the coal and boiler firing conditions, so inlet  $\text{SO}_2$  concentrations fluctuate. Generally, spray dryer inlet  $\text{SO}_2$  concentrations were between 2200 and 2800 ppmv during high-sulfur-coal testing, and 1400 - 1800 ppmv during the medium-sulfur-coal tests.

In the spray dryer, the flue gas contacts a slurry containing slaked lime and recycled solids. A rotary atomizer is used to produce fine slurry droplets. Calcium hydroxide from the lime reacts with flue gas  $\text{SO}_2$  to form calcium sulfite and calcium sulfate solids. The water in the slurry evaporates, cooling and humidifying the hot flue gas. A portion of the product solids drops out of the gas stream and exits through the bottom of the spray dryer. Treated flue gas and the remaining solids go to the ESP, where most of the entrained solids are removed from the gas stream.

All product solids from the spray dryer and ESP hoppers are conveyed to a storage silo, then reslurried. A portion of the reslurried solids is used as a recycle stream in the atomizer feed. In most utility spray dryer systems, excess solids that are not recycled are sent to disposal as a dry powder. However, since a large waste pond is conveniently located in this pilot facility, it is easier to slurry all of the solids and pump the excess to the pond.

Fresh lime slurry is prepared by slaking Mississippi Lime pebble quicklime in a Dixie-Cahaba slaker. Separate slurries of 25 weight percent fresh lime and 35 weight percent recycle solids are pumped to the top of the spray dryer, then combined just before being fed to the atomizer.

### PILOT UNIT EQUIPMENT

The spray dryer vessel is 19.5 ft in diameter with an 11.75 ft straight side. It has a 60° cone bottom, and a side gas exit design. While treating approximately 35,000 acfm of flue gas at spray dryer inlet conditions (30,300 acfm at outlet conditions), the vessel has a flue gas residence time of 10.0 seconds.

The spray dryer is equipped with a rotary atomizer driven by a 100 hp electric motor. The motor has a speed controller which regulates frequency and voltage to control the wheel speed between 7,500 and 13,000 rpm. With a 320 mm (12.6

inch) atomizer wheel diameter, this corresponds to a wheel tip speed range of 412 to 715 ft/sec. The wheel has four 8 mm (5/16 inch) diameter nozzles.

The ESP construction is of rigid frame design with self-tensioning spiral wire electrodes. The ESP has 8 gas passages with a plate height of 23 feet and a spacing of 10 inches, and an aspect ratio (effective length divided by effective height) of 1.6. At the design flue gas flow rate of 35,000 acfm, the specific collection area (SCA) is 387 ft<sup>2</sup>/kacfm, and the face velocity is 3.8 ft/sec. Four electric fields have transformer-rectifiers rated for 200 ma at 55 kV. The housing has room for a fifth field, but there are no internals and the floor is flat in this empty chamber. Rapping is by tumbling hammers activated by an adjustable microprocessor.

## RESULTS

### SO<sub>2</sub> REMOVAL

Statistically-designed experimental programs were conducted to determine SO<sub>2</sub> removal efficiency as a function of major spray dryer operating variables for three different coals. The variables included spray dryer inlet gas temperature, approach to adiabatic saturation, and reagent ratio. Reagent ratio is defined as the moles of calcium in the fresh lime per mole of SO<sub>2</sub> in the spray dryer inlet flue gas, and does not consider any remaining lime content in the recycled solids. All tests were conducted using slaked Mississippi lime reagent. Three Western Kentucky coals were fired including a high-sulfur, low-chloride coal, a high-sulfur, high-chloride coal, and a medium-sulfur, high-chloride coal.

For most of the high-sulfur-coal testing, the high-sulfur, low-chloride coal was fired in the flue gas source boiler. This was Warrior coal from Western Kentucky seam #9, a washed coal with a sulfur content of 4.0% and a chloride content of 0.05%. In December 1987, an unwashed coal with a similar 4.0% sulfur content but higher chloride content of 0.25% was fired. This coal also came from Western Kentucky seam #9 but was purchased from the Pyro Coal Co. Medium-sulfur coal with a sulfur content of 2.5% and a chloride content of 0.25% has been fired since January 1988. The medium-sulfur coal is purchased from the Pyro Coal Co. and is a blend consisting of 85% washed Western Kentucky seam #9 high-sulfur coal and 15% unwashed low-sulfur coal.

#### High-Sulfur, Low-Chloride Coal

The experimental range for the major spray dryer operating variables for the high-sulfur, low-chloride coal tests included spray dryer inlet gas temperatures of 290°F and 320°F, approach temperatures from 18°F to 38°F, and reagent ratio values from 1.0 to 1.6. The full factorial experimental test design consisted of 36 tests with 2 tests at each set of operating conditions.

Figure 2 illustrates SO<sub>2</sub> removal performance data for the spray dryer, the ESP, and the overall system at a spray dryer inlet temperature of 320°F and an approach temperature of 18°F. The data are presented as percent SO<sub>2</sub> removal versus reagent ratio. Three curves are illustrated in each graph, where the top curve represents overall system SO<sub>2</sub> removal performance, the curve just below it represents the percent removal across the spray dryer vessel, and the bottom curve represents the contribution of the ESP to overall SO<sub>2</sub> removal. This ESP removal contribution is calculated by dividing the amount of SO<sub>2</sub> removed in the ESP by the system inlet SO<sub>2</sub> concentration, not by the ESP inlet SO<sub>2</sub> concentration. Thus the ESP contribution can be added directly to the spray dryer contribution to calculate the overall SO<sub>2</sub> removal at a given reagent ratio.



The ESP contribution to overall system SO<sub>2</sub> removal for the conditions illustrated in Figure 2 is typical of the ESP contribution observed at most other conditions, including for other coal types. Generally, the ESP contributed approximately 8 to 14 percentage points to overall system SO<sub>2</sub> removal, with a small dependence on reagent ratio. This is somewhat less removal than has been observed across a fabric filter under similar conditions (3).

Figure 3 illustrates overall system SO<sub>2</sub> removal performance at spray dryer inlet temperatures of 290°F and 320°F and approach temperatures of 18°F, 28°F and 38°F. The approach temperature, or approach to adiabatic saturation at the spray dryer outlet, has a strong influence on SO<sub>2</sub> removal performance while system inlet temperature has a weaker influence. Although SO<sub>2</sub> removal is improved by operating at a low approach to adiabatic saturation, very low approach temperatures may lead to accumulation of wet deposits in the spray dryer vessel. In large, high-sulfur-coal applications where reagent costs represent a significant portion of the system operating expense, the approach to adiabatic saturation must be controlled as low as possible without causing buildup of wet deposits. Common industry practice is to avoid approach temperatures lower than approximately 18°F to 25°F.

The overall system SO<sub>2</sub> removal curves shown in Figures 2 and 3, and the spray dryer SO<sub>2</sub> removal curves shown in Figure 2 are based on multiple regression analyses for the results from 36 tests in the experimental plan. The curves are fitted to the data set as a whole, so the fit through some of the 6-test subsets of the data at a given inlet temperature and approach to adiabatic saturation (e.g., 290°F inlet and 28°F approach as one such subset) may not seem very good. However, while a curve based on a simple regression of 6 test results at a given inlet and approach temperature, with reagent ratio as a variable, would provide a better fit, it would not be as versatile as one multiple regression equation. Furthermore, the one multiple regression would have to be replaced with 6 individual regression curves that could not be used to estimate performance at intermediate values for inlet temperature and approach temperature. Finally, the one multiple regression equation does provide a good fit to the data; it fits all of the 36 data points for overall removal within  $\pm 5$  percentage points.

It should be emphasized that the regression is valid only for this particular coal within the range of operating conditions tested, and it should not be used for coals with different sulfur or chloride contents. The regression equation for overall system SO<sub>2</sub> removal is:

$$\begin{aligned} \% \text{ Overall SO}_2 \text{ Removal} = 100 - e^{\{3.54 + [0.0295 (A-28)] \\ - [0.638 (R-1.3)] \\ - [0.00372 (T-305)] \\ + [0.0345 (A-28) (R-1.3)] \\ - [0.000898 (A-28)^2]\}} \end{aligned} \quad (\text{Eq. 1})$$

where: A = approach-to-adiabatic saturation, °F;  
R = reagent ratio, moles Ca per mole SO<sub>2</sub>; and  
T = flue gas inlet temperature, °F.

The measured overall system SO<sub>2</sub> removal varied from 50% at a 1.0 reagent ratio, 38°F approach, and 290°F inlet, to 85% at a 1.6 reagent ratio, 18°F approach, and 320°F inlet. An overall SO<sub>2</sub> removal efficiency of 90% was not achieved for the high-sulfur, low-chloride coal within the range of operating conditions tested. Spray dryer removal levels varied from about 44% to 72%, while the ESP contribution to overall removal varied from about 2 to 14 percentage points. This ESP contribution to overall removal corresponds to actual percent

SO<sub>2</sub> removal levels across the ESP (based on the ESP inlet SO<sub>2</sub> concentration) of 4 to 45%.

#### Effect of Coal Chloride on SO<sub>2</sub> Removal

Near the end of the high-sulfur coal phase of the test program, a small amount of coal was purchased from another source. The alternate coal had a similar sulfur content of 4.0%, but a higher chloride content of 0.25%. To quantify the effects from the coal change, an abbreviated test plan consisting of ten tests was implemented. The new plan covered the same range of major operating variables as used for the low-chloride coal tests, although not every test condition was used and no tests were replicated.

Figure 4 illustrates overall system SO<sub>2</sub> removal results from the high-sulfur, high-chloride coal tests. Overall SO<sub>2</sub> removal improved by about 8 to 10 percentage points with the change in coal. For a 320°F spray dryer inlet temperature and an 18°F approach to adiabatic saturation, overall SO<sub>2</sub> removals of 80% and 93% were obtained at reagent ratios of 1.0 and 1.6, respectively. Figure 4 indicates that 90% SO<sub>2</sub> removal can be achieved at a reagent ratio of 1.4 while firing this coal.

Chloride content was the most obvious difference between the two coals, and provides the most plausible explanation for the difference in SO<sub>2</sub> removal performance. The alkalinity of the ash from both Western Kentucky coals was low, and is not believed to have been a factor. Chloride in the coal produces HCl in the flue gas which reacts with calcium hydroxide in the spray dryer to form calcium chloride. Calcium chloride has been reported in previous literature (4, 5, 6, 7, and 8) to significantly improve SO<sub>2</sub> removal at a given reagent ratio in spray dryer FGD systems due to its deliquescent nature. The term deliquescent refers to a species which absorbs moisture (is hygroscopic) and is soluble in the absorbed moisture. In the spray dryer, this property can enhance SO<sub>2</sub> removal as it extends the time for aqueous phase reactions to proceed in the droplets. In the ESP, the deliquescent property results in higher residual moisture levels in the nearly dry solids. This increased moisture level is thought to enhance the rate of reaction between flue gas SO<sub>2</sub> and the solids.

Previous test results showing the effects of chloride were obtained by spiking the atomizer feed slurry with calcium chloride. Coal chloride has been recognized as a natural source for increased chloride levels (4, 6, 7). However, results from prior research generally have been presented as a function of chloride level in the spray dried solids, and these levels were not related to specific coal chloride content. Recycle solids chloride levels of 0.2% were obtained while firing the low-chloride coal, and chloride levels of about 0.9 to 1.1% were obtained in the recycle solids while firing the high-chloride coal. The lower of these values corresponds to levels where little, if any, SO<sub>2</sub> removal benefits have been observed previously, while the higher value corresponds to a chloride level where substantial benefits have been previously reported.

Most previously reported spray dryer SO<sub>2</sub> removal results are for flue gas from low-sulfur, low-chloride western coals, so coal chloride has had little impact on those results. The results from this program demonstrate that coal chloride content can directly increase atomizer feed slurry chloride content to levels that previously have been determined to enhance SO<sub>2</sub> removal performance. Furthermore, these increased chloride levels resulting from the coal chloride content appear to have markedly improved system SO<sub>2</sub> removal performance. This underlines the importance of coal chloride content as a variable in determining high-sulfur spray dryer FGD performance.



Increases chloride levels in a spray dryer also may have some detrimental effects. Extension of droplet drying time may increase tendencies for deposition of wet solids on the spray dryer vessel. In this program, increased vessel deposits were observed after switching to the high-chloride coal. Spray dryer operation is discussed later in this paper. Also, chloride may accelerate corrosion, especially stress corrosion in stainless steel parts such as fabric filter bag clamps (4). Finally, chloride may reduce the compressive strength and increase the permeability of solid waste material (4, 9).

#### Effect of Inlet SO<sub>2</sub> Concentration on SO<sub>2</sub> Removal

A third coal fired was a medium-sulfur, high-chloride coal which contained 2.5% sulfur and 0.25% chloride. The effect of inlet SO<sub>2</sub> concentration on SO<sub>2</sub> removal performance can be determined by comparing the SO<sub>2</sub> removal results from this coal with the results from the high-sulfur, high-chloride coal. The major operating variables covered during testing with this coal included inlet gas temperatures of 260°F, 290°F, and 320°F; approach temperatures of 18°F and 28°F; and reagent ratios of 1.0 to 1.6. Each set of test conditions in the factorial test plan was run once, resulting in a total of 18 test runs.

Figure 5 illustrates SO<sub>2</sub> removal results for the medium-sulfur, high-chloride coal. The measured overall system SO<sub>2</sub> removal efficiency varies from 74% at a 1.0 reagent ratio, 28°F approach, and 290°F inlet, to 99% at a 1.6 reagent ratio, 18°F approach, and 320°F inlet. Overall SO<sub>2</sub> removal efficiencies greater than 90% were achieved at a reagent ratio of 1.3 and an 18°F approach temperature.

The increased SO<sub>2</sub> removal efficiency resulting from a decrease in inlet SO<sub>2</sub> concentration is attributed to two causes. First, to achieve the same percentage SO<sub>2</sub> removal at lower concentrations, less SO<sub>2</sub> must be transferred across nearly the same droplet surface area, less lime must dissolve into essentially a constant amount of water, and less calcium sulfite and calcium sulfate must precipitate. The amount of water required to cool the flue gas does not change appreciably with SO<sub>2</sub> concentration, although there is a slight difference attributable to the heat of reaction between SO<sub>2</sub> and lime. Second, less lime is required to achieve a given reagent ratio at lower inlet SO<sub>2</sub> concentrations, so more recycle solids can be added to the feed slurry. Therefore, the opportunity to utilize unreacted lime from the recycle solids is increased. Both of these effects would tend to promote improved SO<sub>2</sub> removal as the spray dryer inlet SO<sub>2</sub> level is reduced.

Current plans are to fire a medium-sulfur, low-chloride coal in 1989, and to spike the recycle solids with CaCl<sub>2</sub> during some tests to simulate the effects of a medium-sulfur, medium-chloride coal. A total of three coal chloride contents for medium-sulfur coal will be tested to determine whether the effects of chloride content on SO<sub>2</sub> removal are non-linear. This will provide data for an overall regression equation which will include coal sulfur and chloride contents as variables.

#### ESP PARTICULATE REMOVAL

To quantify the effects of a spray dryer FGD retrofit on an existing ESP, operation of the pilot ESP was evaluated with and without the spray dryer in service. Five tests were made without spray dryer operation to characterize baseline ESP particulate removal for normal fly ash. SCA and gas velocity were varied from test to test by de-energizing ESP electrical fields and varying flue gas flow rates. Later, five comparable tests were made with the spray dryer in operation. For those tests, the reagent ratio was set at 1.3, the approach temperature was controlled at 20°F, and recycle solids were fed as a separate slurry containing 35

weight percent solids. Particulate removal results for the two sets of five tests are summarized in Table 1.

To simulate the changes that occur after a spray dryer retrofit, the spray dryer inlet gas conditions in the second set of tests were identical to the ESP inlet gas conditions for the baseline (fly ash only) runs. Thus, the treated gas going to the ESP was more humid, cooler, and at a lower volumetric flow rate in the second set of tests. This lowered the ESP gas velocity and increased the SCA, realistic benefits that would result from retrofitting a spray dryer. In Table 1, the flue gas flow rate for condition 1 with the spray dryer off corresponds to that for condition 1 with the spray dryer on, after the change in flue gas temperature and moisture content effected by the spray dryer are considered.

Table 1 shows that the ESP inlet mass loading with the spray dryer off ranged from 0.6 to 1.2 grains per actual cubic foot (gr/acf), then increased to about 9 to 10 gr/acf with the addition of the spray dryer. Thus, the spray dryer upstream increased the ESP inlet mass loadings by approximately a factor of 10. However, ESP outlet mass loadings also increased by approximately that same factor. Therefore, ESP particulate removal efficiencies were about the same before and after addition of the spray dryer, but outlet emission rates increased significantly. With the exception of a low SCA test and a high gas velocity test, however, the emission rates were still below the Shawnee plant emission limit of 0.11 lb/10<sup>6</sup> BTU as set by the State of Kentucky.

The lack of improvement in particulate removal efficiency after addition of the spray dryer is surprising for several reasons. First, cooling of the flue gas reduces the volumetric flow rate, which produces the benefits of increased SCA and reduced gas velocity, as mentioned above. Second, the majority of the increase in mass loading across the spray dryer is greater than 5 micrometers in diameter and should be easily collected by an ESP. Third, at the lower temperatures with the spray dryer in service, the gas viscosity is decreased, allowing particles to more readily migrate to the collecting plates. Finally, higher electrical field strengths are obtainable after addition of the spray dryer. This is because the cool spray dryer exit gas is more dense than flue gas at normal temperatures, and electrical breakdown in the gas space between electrodes is greatly reduced at higher gas density. As an example of this effect, without the spray dryer in service the first field of the ESP was limited by sparking to an operating voltage of less than 40 kV, and an operating current density of about 30 nA/cm<sup>2</sup>. With the spray dryer in service, voltages of nearly 50 kV and current densities of 65 nA/cm<sup>2</sup> were obtained in the first field. These improvements in electrical operation increased the electric field strength by nearly 30%. With the spray dryer in operation, electrical field strengths were limited by the maximum current output from the transformer-rectifier sets rather than by the onset of electrical breakdown in the gas space.

Furthermore, two potential detrimental effects of adding a spray dryer upstream of an ESP were not observed. One potential detrimental effect would be a result of the high particulate concentrations downstream of the spray dryer. An increase in fine particulate mass loading can reduce the voltage gradient near the emitting electrodes, resulting in a space charge quenching that is sufficient to suppress corona formation. The formation of a corona, or heavily ionized gas, at the emitting electrodes is an important step in the process by which particles are charged. However, the increased mass was concentrated in larger particle size intervals, and no space charge quenching was detected.

A second detrimental effect anticipated was an increase in ash resistivity. Most high-sulfur coals produce a fly ash with a low resistivity, due to conditioning by the SO<sub>3</sub> present in the flue gas. This SO<sub>3</sub> presumably is removed by the

spray dryer, thus negating this conditioning effect. Furthermore, the calcium-based salts added by the spray dryer have a high inherent resistivity ( $>10^{12}$  ohm-cm). However, rather than being increased, particulate resistivity was decreased as a result of the added moisture and reduced flue gas temperature.

Because theory would predict improvement in ESP particulate removal efficiency due to the higher electrical field strengths, increased particle size, and other benefits due to humidification and cooling after the addition of the spray dryer, the lack of improvement in efficiency is likely due to non-ideal effects. An examination of particle size distribution data from these tests reveals that the increase in mass loading at the ESP inlet (or spray dryer outlet) is mostly due to particles that are 5 to 20 micrometers in physical diameter, and that these particles were removed very efficiently by the ESP. The lack of improvement in particulate removal efficiency during spray dryer operation is mostly due to decreased removal efficiency for particles that have a physical diameter of 2 to 5 micrometers. This is surprising because the spray dryer does not significantly increase the mass loading for these particle sizes. This observation points to non-ideal effects in general, and the magnitude of the emissions suggests that re-entrainment (e.g., rapping or scouring) in particular, is the cause of this limitation on ESP performance.

Further evidence of re-entrainment is seen in comparing the results of spray dryer operation tests at condition 3 to those at condition 2. At condition 3, with a flue gas velocity of 4.1 ft/sec, the ESP particulate removal efficiency is lower than that of condition 2, at a velocity of 3.6 ft/sec. This lowered particulate removal efficiency occurs in spite of condition 3 having a slightly higher SCA value. The fact that increased gas velocity caused a reduced particulate removal efficiency, whereas ESP theory says that velocity should have no effect, points to a non-ideal effect such as re-entrainment being the major limitation on ESP performance. Resolution of this indicated problem is being investigated. More detailed discussion of the ESP particulate removal test results is provided in two previous papers (10, 11).

#### SPRAY DRYER VESSEL DEPOSITS

Deposits of damp solids within the spray dryer vessel were a significant problem, particularly in the earlier stages of pilot unit operation. It should be noted that since this was a research program, the test conditions called for in the test plan sometimes proved to be outside the range over which the spray dryer could operate successfully. By attempting conditions which resulted in poor drying, though, it was possible to better define operating limits for the system.

The spray dryer bottom plugged with damp solids on several occasions. This occurred during instances when the walls and ceiling of the spray dryer vessel had a large amount of solids build-up. Large chunks of solid material would dislodge from the walls and ceiling intermittently and fall into the vessel bottom. Ceiling deposits would grow to be 6 to 12 inches thick and then an area of several square feet would break loose. This would plug the opening in the vessel bottom, or the opening of the lump breaker which was installed in an effort to reduce the size of these fallen agglomerations. Wall deposits tended not to slough off, but continued to accumulate until they became 2 to 3 feet thick and cleaning was deemed necessary. The spray dryer was generally inspected every 1 to 2 weeks, depending on operating conditions, to monitor deposit accumulation.

The deposits were attributed to conditions which, when combined, resulted in poor drying. These conditions included low approach temperatures, low wheel tip speed, low solids content in the atomizer feed slurry, high coal chloride content,



and low spray dryer residence time. Table 2 summarizes the combinations of conditions which were inoperable (deposits quickly plugged the spray dryer bottom), operable but troublesome (some deposits buildup and plugging, but tests could be completed), and operable (no significant deposits buildup). Not all combinations of wheel tip speed, feed slurry solids content, coal chloride content, and spray dryer residence time were tested. Table 2 summarizes results of the combinations of those variables that were tested. Specifics of the spray dryer vessel design, such as the design of the flue gas distributor, might also have influenced the effectiveness of droplet drying. However, vessel design effects could not be evaluated as part of the test program.

One variable in Table 2, adjustment for sootblowing, requires some explanation. About one third of the tests were conducted at an 18°F approach to adiabatic saturation. For the first portion of this test program, steam sootblowing in the flue gas source boiler resulted in very low approach temperatures during these tests for approximately one to four hours per day. Steam sootblowing adds a significant amount of moisture to the flue gas, and causes wet bulb temperatures to rise by as much as 5 to 6°F. Because the spray dryer outlet temperature was not raised by a like amount, the approach temperature was effectively lowered from 18°F to 12°F. A 12°F approach to adiabatic saturation was not proven to be viable under any combination of conditions.

Spray dryer FGD systems for high-sulfur-coal applications will likely have to operate at relatively low approach temperature values to achieve required SO<sub>2</sub> removal levels. Utilities that operate such systems at approach temperatures of 18 to 20°F would certainly have to adjust the spray dryer outlet temperature during steam sootblowing. To more closely reflect utility operating practices, a steam flow rate signal was recently added at the spray dryer control room to indicate when sootblowing occurs, and adjustments to the spray dryer outlet temperature control are now being made.

During preliminary atomizer speed screening tests, atomizer tip speed was found to have a significant effect on vessel deposits. Ceiling and wall deposits and plugging of the spray dryer bottom were problems at an atomizer speed of 8500 rpm (tip speed of 467 ft/sec) even at a 38°F approach temperature. Operation at a higher speed of 10,500 rpm (tip speed of 577 ft/sec) but a lower approach temperature of 17°F also was inoperable. The spray dryer bottom plugged repeatedly with fallen deposits.

Several tests were successfully completed at 11,500 rpm (tip speed of 632 ft/sec) and an 18°F approach temperature, although the buildup of deposits was significant. Some runs were interrupted by plugging of the spray dryer bottom, but were finished after cleaning. On one occasion, very large deposits were found on the spray dryer walls. The deposits had grown out from the wall, between 1 to 5 feet below the ceiling, toward the atomizer wheel. The deposits were 6 inches to 2 feet thick all around the wall. Approximately 10 larger growths, 1 to 2 feet in diameter, projected another 2 to 3 feet toward the atomizer wheel. The deposits were removed using a high pressure water hose. All of the tests mentioned above were conducted before pilot unit operation was changed to raise the spray dryer outlet temperature during boiler soot blowing.

Later, the atomizer speed was increased to 12,900 rpm (tip speed of 709 ft/sec). Although both 632 ft/sec and 709 ft/sec are within the range of tip speeds for full-scale utility spray dryer rotary atomizers, operation of this system was much better at the higher value. After the speed was increased, the tendency for wall deposits to accumulate decreased significantly for most test conditions.



However, some conditions still resulted in solids accumulation and spray dryer bottom plugging. One set of conditions which increased deposits was once-through operation at reagent ratios between 1.0 and 1.6. In these tests, lime slurry was fed to control reagent ratio and plain water (no recycle) was fed to control the approach to adiabatic saturation. The solids content of the atomizer feed was only about 12 weight percent. However, none of these tests were successfully completed, due to excessive deposit formation in the spray dryer. This is attributed to the low solids content of the slurry. Slurry droplets with a low solids content take longer to dry than those with a higher solids content. It is unlikely that a full-scale installation would operate in a once-through configuration at a low (18°F) approach to adiabatic saturation. Due to the separate slurry recycle preparation method used in this pilot unit, mixing a 25 weight percent lime slurry with a 35 weight percent recycle slurry, even typical runs had a relatively low atomizer feed solids content of about 30 weight percent. An atomizer feed solids content of 40 weight percent is achievable using a combined lime and recycle slurry feed method, or by going to higher solids levels in the recycle slurry. This would help full-scale systems avoid accumulation of deposits.

The rate of deposits accumulation increased when high-chloride (0.25% Cl) coal was fired. This is attributed to the deliquescent nature of calcium chloride, which would be expected to increase the droplet drying time and which caused the product solids to have a higher residual moisture content. The plugging problems associated with the increased coal chloride level were virtually eliminated, though, by adjusting the spray dryer outlet temperature during steam soot blowing.

Spray dryer vessel geometry and residence time establish the time available for a droplet to dry before it contacts the vessel walls. In this program, a spray dryer residence time of 10 seconds was used for most of the tests. This is at the lower end of the range of design residence times for full-scale spray dryer FGD systems.

The system operated successfully at a spray dryer residence time of 10 seconds, an atomizer tip speed of 709 ft/sec, an 18°F approach to adiabatic saturation (with adjustments made to the spray dryer outlet temperature during soot-blowing), a 30 weight percent solids content in the atomizer feed slurry, and a 0.25 weight percent coal chloride content. However, when the gas flow rate was increased to reduce the spray dryer residence time to 9.2 seconds, plugging of the spray dryer bottom frequently occurred.

A full-scale system should be able to minimize recurring deposition problems through system design and selection of operating conditions. The design must ensure sufficient vessel flue gas residence time, atomizer tip speed, and feed slurry solids content to produce adequate drying within the spray dryer. Also, coal chloride content should be considered in establishing design and operating conditions. Furthermore, a full-scale system must take measures to ensure that conditions such as steam sootblowing in the boiler do not lower the approach temperature below the design value. Because there are several interrelated factors which affect the rate of deposits accumulation as well as SO<sub>2</sub> removal efficiency, specific levels for each factor cannot be recommended here. Further work is needed to establish optimized combinations for these factors, particularly over a range of coal chloride levels.

#### SUMMARY

After one-and-a-half years of testing for two high-sulfur coals and one medium-sulfur coal at TVA's 10-MW spray dryer/ESP pilot plant, several observations may be made about the performance of spray dryer/ESP technology. SO<sub>2</sub>

removal efficiencies greater than 80% are readily achieved while firing high-sulfur, low-chloride coal. Under some conditions, greater than 90% SO<sub>2</sub> removal may be achieved, but high reagent ratios would be required and this may not be economical. Increasing the chloride content and decreasing the sulfur content of the coal result in improved SO<sub>2</sub> removal efficiency. Ninety percent SO<sub>2</sub> removal efficiency can be achieved at a reagent ratio of 1.3 while firing medium-sulfur, low-chloride coal. Although the effect of chloride was observed with a change in the chloride content of the coal, previous studies have shown that a similar effect is obtainable by spiking recycle solids with calcium chloride (4, 5). Table 3 summarizes the observed reagent ratios required to achieve 90% SO<sub>2</sub> removal, and SO<sub>2</sub> removal efficiencies at a reagent ratio of 1.6 and at an 18°F approach to adiabatic saturation.

The addition of a spray dryer upstream greatly increased the mass loading at the ESP inlet, but also improved the electrical operating characteristics of the ESP. The net effect for this pilot unit was no change in ESP particulate removal efficiency, but a corresponding increase in ESP outlet emissions when the spray dryer was in service. This is believed to be due to non-ideal effects such as scouring or rapping re-entrainment.

A more specific summary of the results is provided below.

#### SO<sub>2</sub> Removal Performance

- For a 4% sulfur, 0.05% chloride coal, an SO<sub>2</sub> removal level of 85% was achieved at a reagent ratio of 1.6 for operation at an 18°F approach to adiabatic saturation and an inlet temperature of 320°F. Ninety percent removal was not achieved within this range of reagent ratios. However, a further increase in reagent ratio may enable higher removals while firing this coal.
- A change to a 4% sulfur, 0.25% chloride coal raised the overall system SO<sub>2</sub> removal level at a reagent ratio of 1.6 to 93%, for operation at an 18°F approach to adiabatic saturation and an inlet temperature of 320°F. A 90% removal level appears to be achievable at a reagent ratio between 1.4 and 1.5.
- A change to a 2.5% sulfur, 0.25% chloride coal raised the overall system SO<sub>2</sub> removal level at a reagent ratio of 1.6 to 99%, for operation at an 18°F approach to adiabatic saturation and an inlet temperature of 320°F. A 90% removal level appears to be achievable at a reagent ratio of 1.3.
- SO<sub>2</sub> removal within the ESP contributed approximately 8 to 14 percentage points to the overall system removal.

#### ESP Performance

- ESP particulate removal efficiency (about 99.7% with 4 fields in operation) did not change significantly after a spray dryer was added upstream. However, ESP inlet mass loadings increased from about 1 gr/acf to nearly 10 gr/acf with the spray dryer in operation. ESP outlet emissions increased by roughly the same factor, but were below emission limits for the State of Kentucky for most tests.

- The increased density of the cooled spray dryer exit gas allowed the ESP to operate at higher electrical field strengths. Field strengths were limited by the maximum current output from the transformer-rectifier sets rather than electrical breakdown in the gas space. There were no particulate resistivity or corona quenching problems. It appeared that a scouring re-entrainment problem related to low particle resistivity limited ESP particulate control efficiency during spray dryer operation, though.

#### Spray Dryer Equipment Performance

- Build-up of deposits in the spray dryer vessel was a significant problem at some test conditions. This was attributed to poor drying conditions which resulted from combinations of: low approach temperatures that were not adjusted to account for steam sootblowing at the boiler, low atomizer tip speeds, low slurry solids content in the atomizer feed slurry, and high coal chloride levels. However, by increasing the atomizer tip speed to 709 ft/sec and adjusting the spray dryer outlet temperature for steam sootblowing, these deposits were avoided even for operation at an 18°F approach to adiabatic saturation and with a 0.25 weight percent coal chloride level. Spray dryer residence time values of greater than 10 seconds, and an atomizer feed slurry solids content greater than 30 weight percent would further minimize these tendencies.

#### ACKNOWLEDGMENTS

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Tennessee Valley Authority  
Electric Power Research Institute

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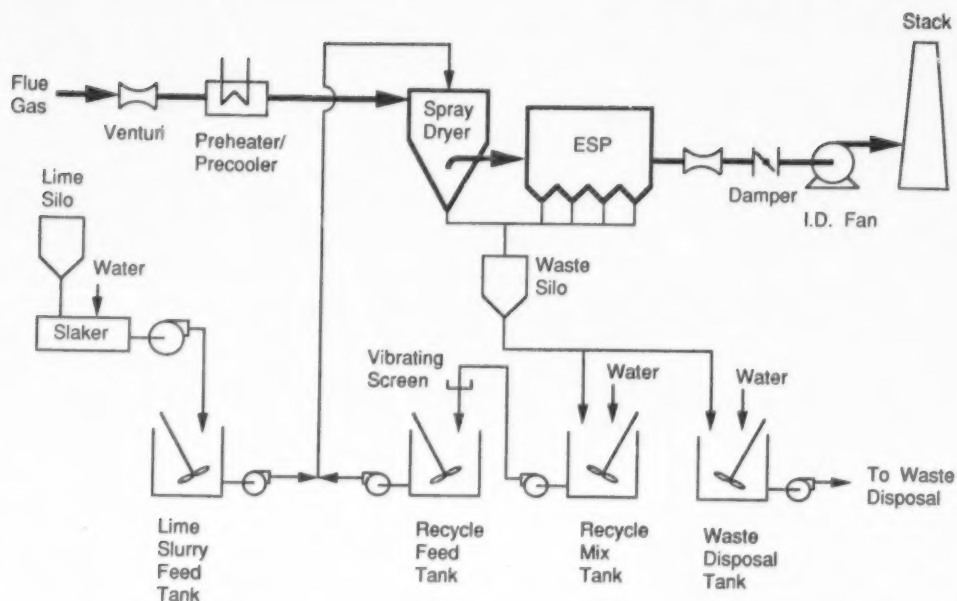


Figure 1. Simplified process flow diagram for TVA pilot spray dryer/ESP.

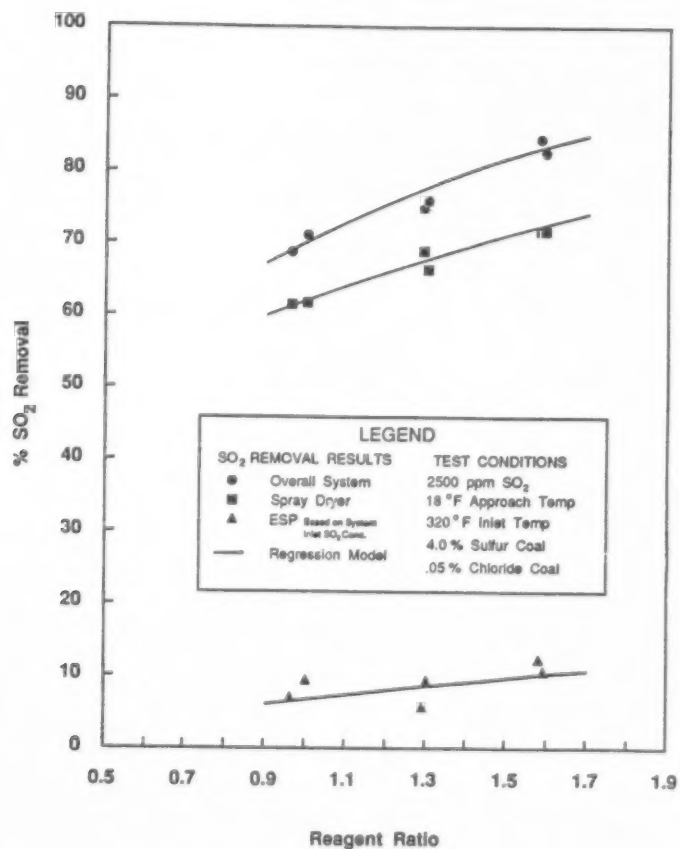
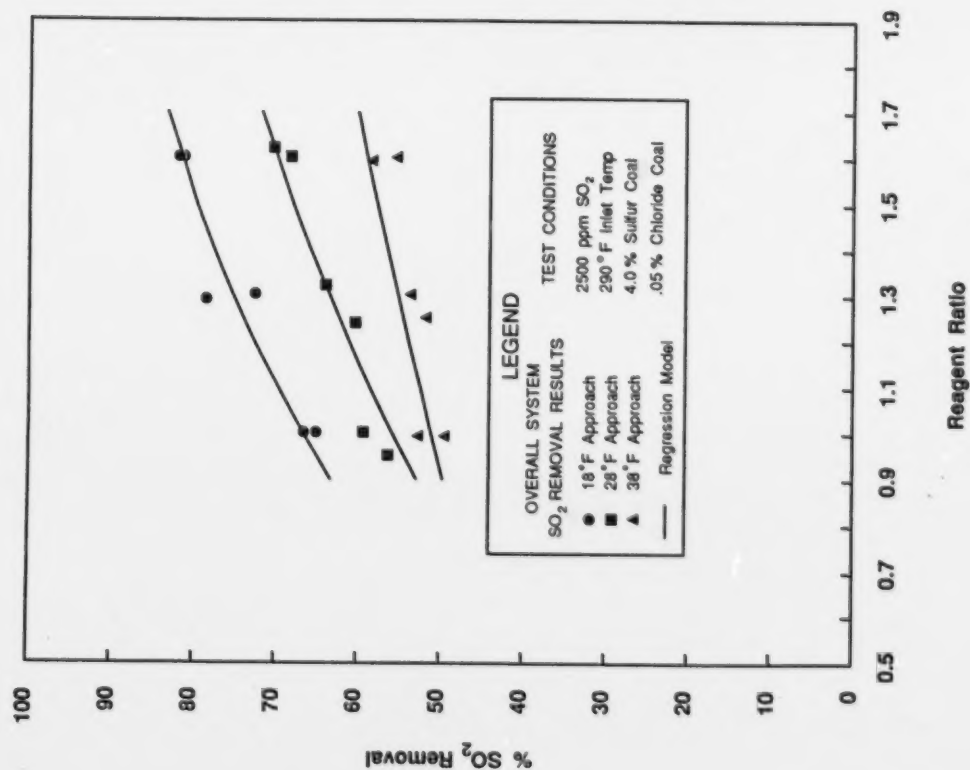
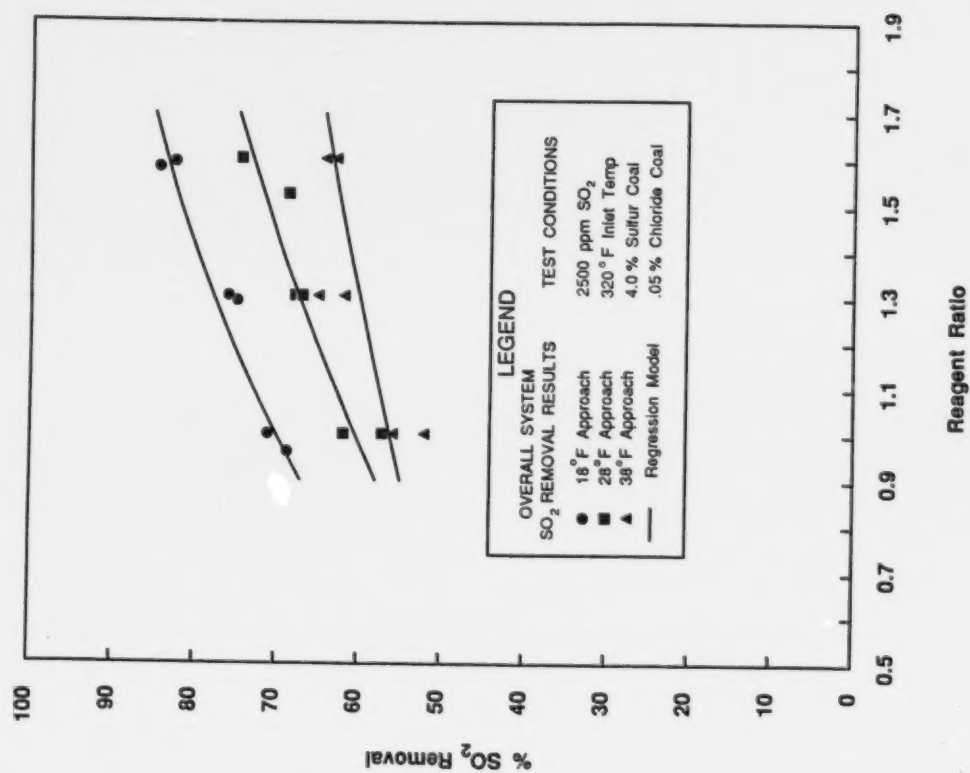


Figure 2. Spray dryer and ESP contributions to overall system SO<sub>2</sub> removal.

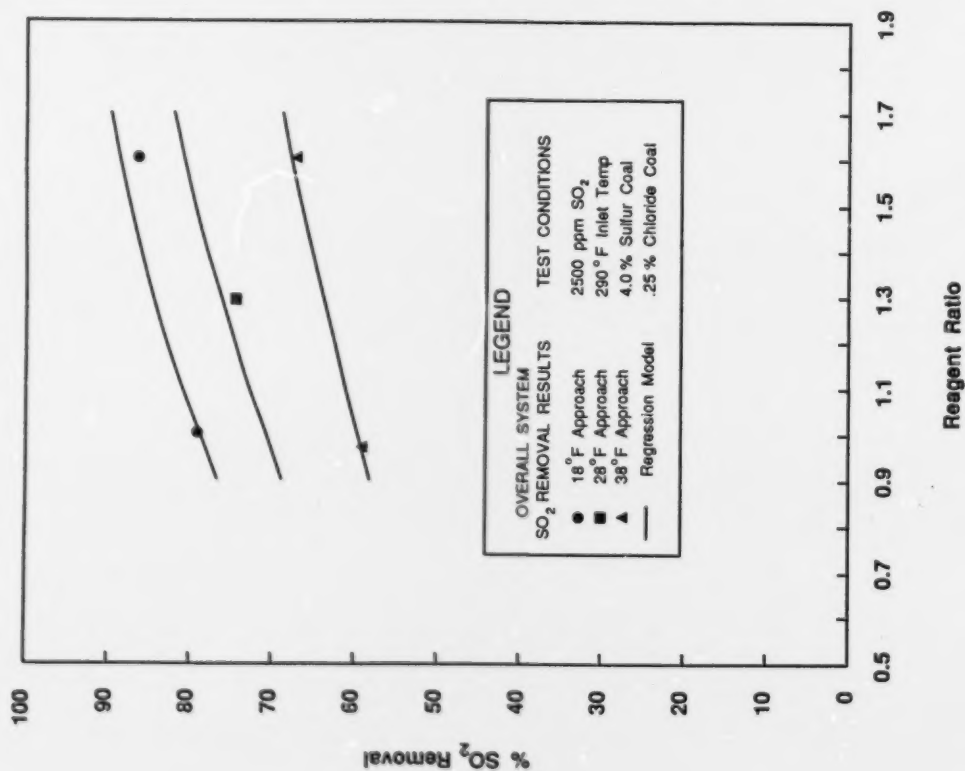


a) 290°F inlet temperature

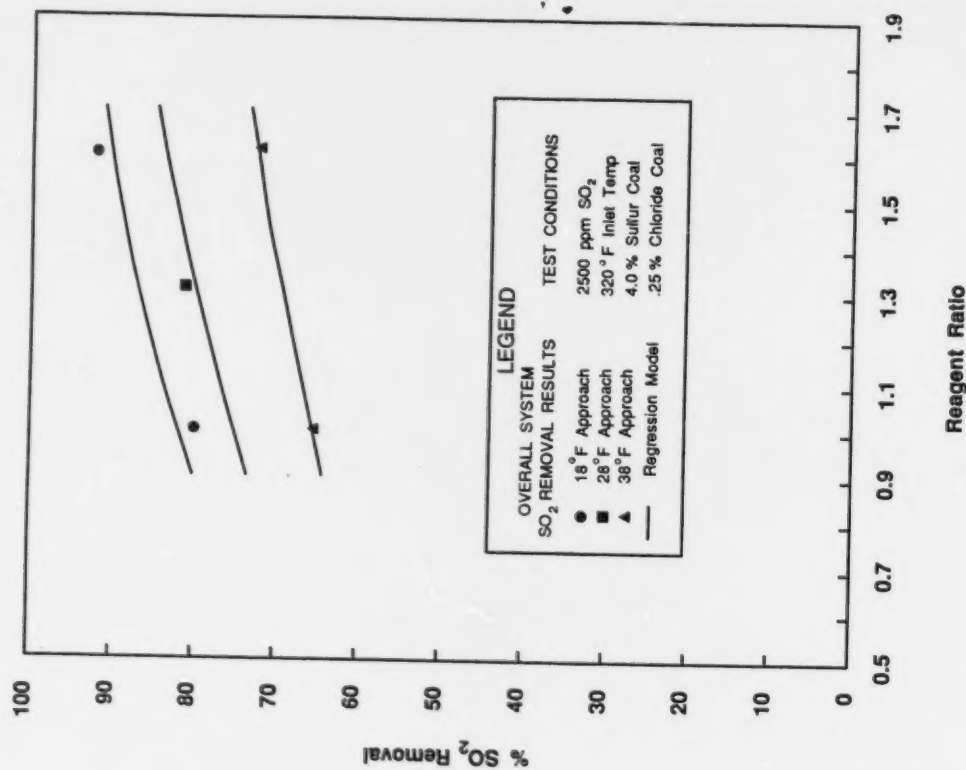


b) 320°F inlet temperature

Figure 3. SO<sub>2</sub> removal performance for high-sulfur, low-chloride coal.

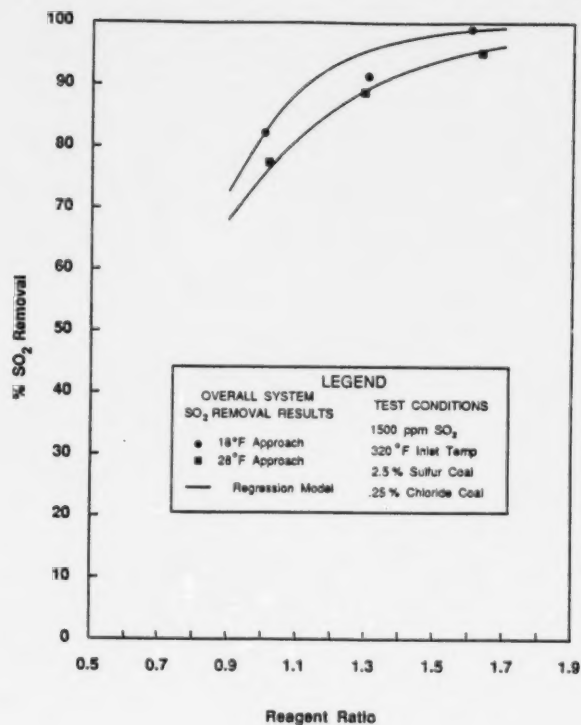


a) 290°F inlet temperature

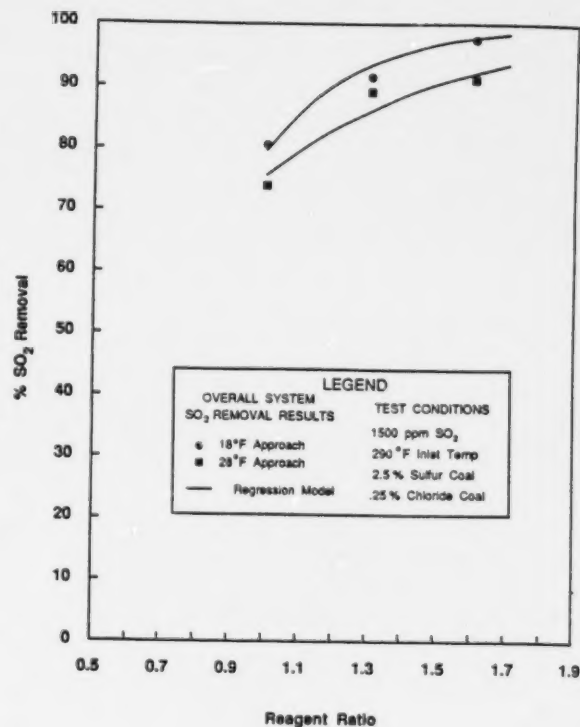


b) 320°F inlet temperature

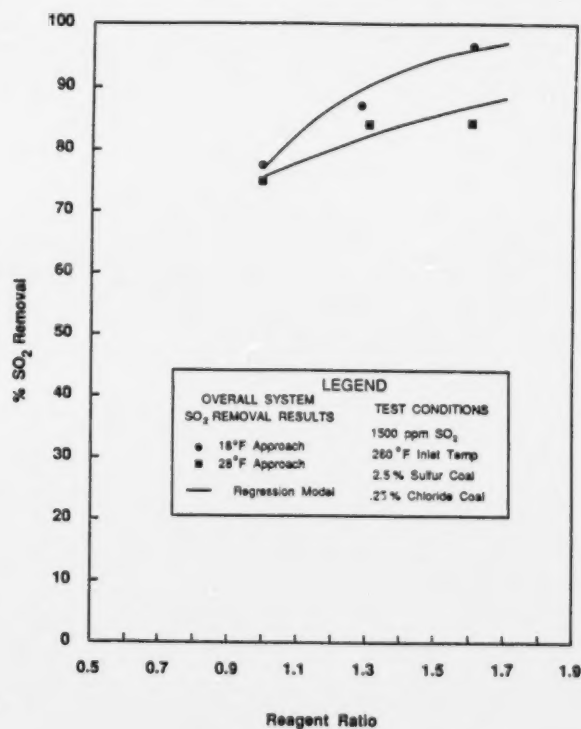
Figure 4. SO<sub>2</sub> removal performance for high-sulfur, high-chloride coal.



a) 260°F inlet temperature



b) 290°F inlet temperature



c) 320°F inlet temperature

Figure 5. SO<sub>2</sub> removal performance for medium-sulfur, high-chloride coal.



TABLE 1. ESP PARTICULATE REMOVAL PERFORMANCE

| Condition       | ESP<br>Inlet<br>Flow, acfm | ESP<br>Inlet<br>Temp.<br>°F | Fields | SCA,<br>sq ft/<br>kacfm | Gas<br>Velocity,<br>ft/sec | Inlet<br>Conc.<br>gr/acf | Outlet<br>Conc.<br>gr/acf | Outlet<br>Conc.<br>gr/dscf | Outlet<br>Conc.<br>lb/10 <sup>6</sup> Btu | Fly<br>Ash<br>Rem.<br>% |
|-----------------|----------------------------|-----------------------------|--------|-------------------------|----------------------------|--------------------------|---------------------------|----------------------------|-------------------------------------------|-------------------------|
| SPRAY DRYER OFF |                            |                             |        |                         |                            |                          |                           |                            |                                           |                         |
| 1               | 34614                      | 319                         | 4      | 390                     | 3.76                       | 1.151                    | 0.00198                   | 0.00332                    | 0.00705                                   | 99.832                  |
| 2               | 34961                      | 317                         | 3      | 290                     | 3.80                       | 0.942                    | 0.00240                   | 0.00404                    | 0.00877                                   | 99.732                  |
| 3               | 43965                      | 317                         | 4      | 307                     | 4.78                       | 0.727                    | 0.00221                   | 0.00380                    | 0.00870                                   | 99.690                  |
| 4               | 26564                      | 318                         | 3      | 381                     | 2.89                       | 0.584                    | 0.00220                   | 0.00372                    | 0.00778                                   | 99.601                  |
| 5               | 35124                      | 316                         | 2      | 192                     | 3.82                       | 0.646                    | 0.00942                   | 0.01582                    | 0.03385                                   | 98.485                  |
| SPRAY DRYER ON  |                            |                             |        |                         |                            |                          |                           |                            |                                           |                         |
| 1               | 32832                      | 142                         | 4      | 412                     | 3.57                       | 9.223                    | 0.0155                    | 0.0219                     | 0.0505                                    | 99.838                  |
| 2               | 32623                      | 144                         | 3      | 311                     | 3.55                       | 8.987                    | 0.0304                    | 0.0433                     | 0.0986                                    | 99.662                  |
| 3               | 37752                      | 145                         | 4      | 358                     | 4.10                       | 10.286                   | 0.0167                    | 0.0893                     | 0.1882                                    | 99.404                  |
| 4               | 24242                      | 144                         | 3      | 419                     | 2.64                       | 9.167                    | 0.0256                    | 0.0355                     | 0.0807                                    | 99.726                  |
| 5               | 31856                      | 147                         | 2      | 212                     | 3.46                       | 8.582                    | 0.2187                    | 0.3134                     | 0.6565                                    | 97.464                  |

Spray Dryer Operating Conditions: Reagent Ratio - 1.3 moles fresh  $\text{Ca(OH)}_2$ /mole inlet  $\text{SO}_2$

Approach to Adiabatic Saturation - 20°F

Inlet Temperature - 320°F

Lime Solids - 25 wt. %

Recycle Solids - 35 wt. %

Inlet  $\text{SO}_2$  - approximately 2500 ppm

TABLE 2. OPERATING CONDITIONS FOR SPRAY DRYER VESSEL DEPOSITS

| Tip Speed, ft/sec                         | Approach, °F | Adjust for Soot-blowing <sup>a</sup> | Coal Chloride | Feed Solids Content | Residence Time, Seconds |
|-------------------------------------------|--------------|--------------------------------------|---------------|---------------------|-------------------------|
| <u>Unoperable Conditions<sup>b</sup></u>  |              |                                      |               |                     |                         |
| 467                                       | 38           | No                                   | .05%          | 30%                 | 10                      |
| 577                                       | 17           | No                                   | .05%          | 30%                 | 10                      |
| 709                                       | 18           | No                                   | .05%          | 12%                 | 10                      |
| <u>Troublesome Conditions<sup>c</sup></u> |              |                                      |               |                     |                         |
| 632                                       | 18           | No                                   | .05%          | 30%                 | 10                      |
| 709                                       | 18           | No                                   | .25%          | 30%                 | 10                      |
| 709                                       | 18           | Yes                                  | .25%          | 30%                 | 9.2                     |
| <u>Operable Conditions<sup>d</sup></u>    |              |                                      |               |                     |                         |
| 709                                       | 18           | No                                   | .05%          | 30%                 | 10                      |
| 709                                       | 18           | Yes                                  | .25%          | 30%                 | 10                      |

<sup>a</sup>Spray dryer outlet temperature increased during steam sootblowing.

<sup>b</sup>Repeated plugging of spray dryer bottom prevented test runs from being completed.

<sup>c</sup>Occasional plugging of spray dryer bottom, but test runs were completed anyway.

<sup>d</sup>No problems with plugging of spray dryer bottom due to deposits in spray dryer vessel.

TABLE 3. SUMMARY OF SO<sub>2</sub> REMOVAL PERFORMANCE CAPABILITIES

| Coal % S | % Cl | Inlet SO <sub>2</sub> , ppm | Inlet Temp, °F | Reagent Ratio Required to Achieve 90% SO <sub>2</sub> Removal (18°F Approach) | % SO <sub>2</sub> Removal at 1.6 Reagent Ratio |
|----------|------|-----------------------------|----------------|-------------------------------------------------------------------------------|------------------------------------------------|
| 4.0      | 0.05 | 2500                        | 320            | >1.6                                                                          | 85%                                            |
|          |      |                             | 290            | >1.6                                                                          | 81%                                            |
| 4.0      | 0.25 | 2500                        | 320            | 1.5                                                                           | 92%                                            |
|          |      |                             | 290            | >1.6                                                                          | 86%                                            |
| 2.5      | 0.25 | 1550                        | 320            | 1.1                                                                           | 99%                                            |
|          |      |                             | 290            | 1.2                                                                           | 98%                                            |
|          |      |                             | 260            | 1.3                                                                           | 97%                                            |

THE GRDA UNIT NO. 2 DRY FGD SYSTEM:  
PERFORMANCE AND OPERATING EXPERIENCE

By: Jack Marcy, Assistant General Manager  
Thermal Generation  
Grand River Dam Authority  
Chouteau, Oklahoma

Tom Lillestolen, Director of Products  
John Buschmann, Product Manager, FGD  
Environmental Systems Division  
Flakt, Inc.  
Knoxville, Tennessee

ABSTRACT

The Grand River Dam Authority (GRDA) Unit No. 2 is a 520-MW coal-fired generating station located near Chouteau, Oklahoma which is designed to burn 0.4-1.5% sulfur coal. The flue gases emitted from the boiler are treated in a Dry FGD System.

The gas cleaning system includes spray dryers, with multiple rotary atomizers, electrostatic precipitators, lime storage, supply and preparation equipment, ash handling, disposal, and recycle equipment.

This paper will review the design of the FGD system and summarize the system performance since beginning commercial operation. Greater than 85% SO<sub>2</sub> removal efficiency and 99.86% particulate removal efficiency have been demonstrated. The high particulate removal has been achieved in spite of the high dust loadings created by the use of recycle.

The operating experience will also be discussed with attention to situations encountered and how they were addressed and solved. Areas cited include recycle slurry handling, lime slaking, slurry abrasive wear of atomizer disks and ash handling.

## INTRODUCTION

The Grand River Dam Authority's (GRDA) Unit No. 2 is a 520-MW coal-fired generating station located near Chouteau, Oklahoma. During the planning stages, it was determined that Unit No. 2 should meet emission standards of the Clean Air Act, including the amendments of 1977. The existing Unit No. 1 only utilized an electrostatic precipitator for particulate control. To satisfy the regulations, it was decided that a flue gas cleaning system should be included in the Unit No. 2 design. It was further decided that the flue gas cleaning system should be comprised of a "dry-scrubber" and an electrostatic precipitator, which could simultaneously remove sulfur dioxide and particulate matter.

GRDA was assisted in this decision and eventual system engineering and design by the Benham-Holway Power Group. GRDA ultimately purchased a Foster Wheeler boiler and a Brown Boveri turbine/generator.

On May 19, 1982, GRDA purchased an Air Quality Control System (AQCS) from Flakt, Inc., Environmental Control Systems Division, for approximately seventy million dollars. The "turn-key" system was complete, including equipment for  $\text{SO}_2$  and particulate control, reagent unloading and storage, ash handling and storage, and all associated process equipment and controls.

An artists conception of the APC System is illustrated in Figure 1.

Flakt constructed the AQCS system during the period from August, 1983 to March, 1985. Equipment start-up commenced immediately and Commercial Operation began in October, 1985. The plant has been operating since then producing electric power on a continuous basis (except for scheduled maintenance periods), exceeding GRDA's expectations.

The following will describe the system and performance requirements for the AQCS system. Furthermore, discussion is provided on the operating experience of the AQCS system and performance results are reported.

## SYSTEM DESCRIPTION

Primary design requirements for the AQCS relate to satisfying the APC Permit, which deal with  $\text{SO}_2$  and particulate emissions. These are summarized in Table 1. Also included in this table are the design values stipulated in the GRDA specification for gas flow, temperature and pressure. Table 1 summarizes the design fuel data for both Wyoming and Oklahoma coals. A special feature of the system design was that the plant should be capable of operating with a blend coal which could contain as much as 1.5% sulfur.

The AQCS system is comprised of spray dryer reactors, with a means for atomizing the reagent solution into small droplets, reactor feed system, recycle system, lime slurry preparation system, dry lime unloading and handling, electrostatic precipitators and fly ash handling. The dry lime unloading and handling system supply lime to the lime slurry preparation system. The lime slurry preparation system prepares a concentrated solution of lime to the reactor feed system. The recycle system returns dried reactor and electrostatic precipitator product back to the reactor feed system after mixing it with water. This is done to minimize the use of lime. The reactor feed system combines feed from the lime slurry preparation system and the recycle system and pumps it to the



FIGURE 1  
PLANT ARRANGEMENT

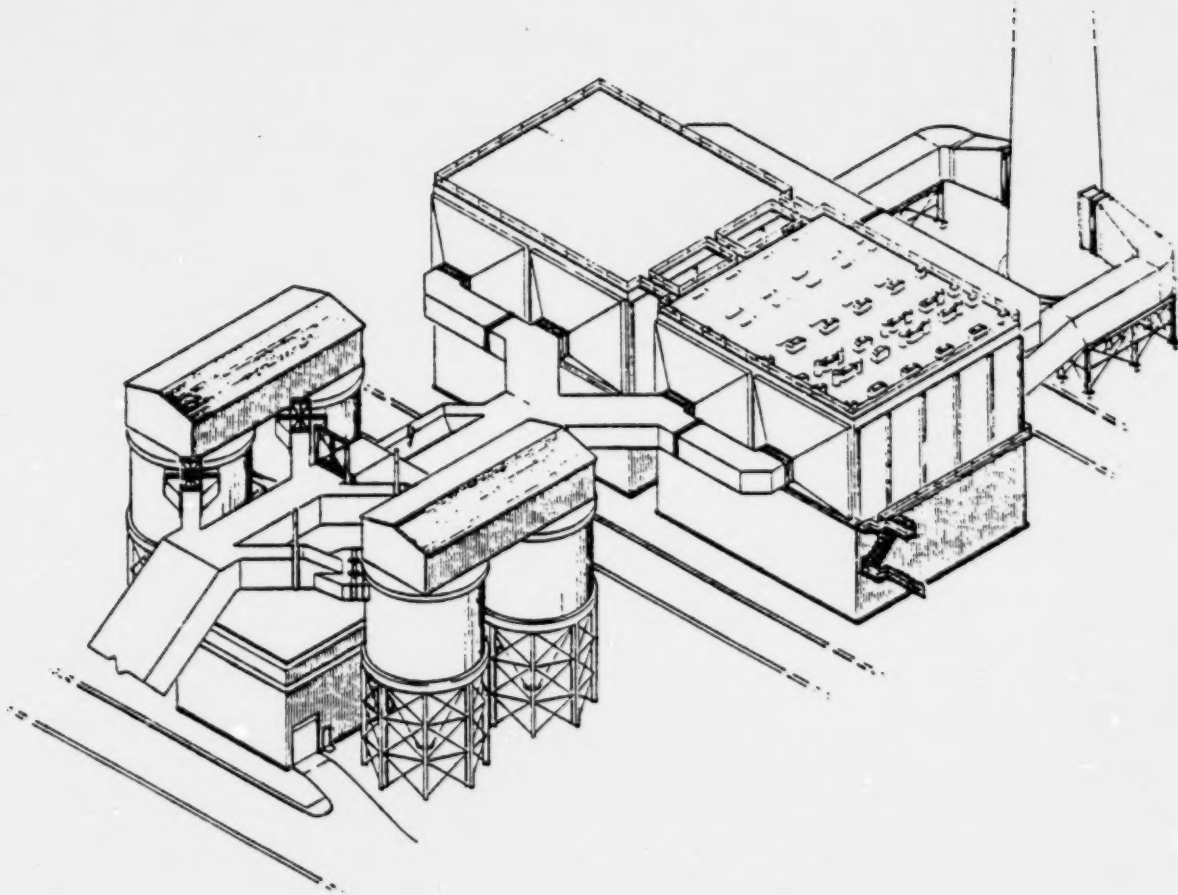


TABLE 1

A. Boiler Data

| Coal Type -<br>Boiler Load:                               | <u>Wyoming Coal</u> |            | <u>50% Blend of Wyoming &amp;<br/>Oklahoma Coal (by Btu)</u> |            |
|-----------------------------------------------------------|---------------------|------------|--------------------------------------------------------------|------------|
|                                                           | <u>100%</u>         | <u>60%</u> | <u>100%</u>                                                  | <u>60%</u> |
| <u>Flue Gas Conditions</u>                                |                     |            |                                                              |            |
| 1. Steam from Super-<br>heater (lb/hr)                    | 3,900,000           | 2,340,000  | 3,900,000                                                    | 2,340,000  |
| 2. Coal Firing Rate,<br>(Tons/hr)                         | 328.74              | 212.45     | 263.43                                                       | 170.89     |
| 3. Economizer Outlet<br>Flue Gas Temp<br>(°F)             | 718                 | 631        | 712                                                          | 635        |
| 4. Air Heater Outlet<br>Flue Gas Flow<br>(lb/hr)          | 5,804,000           | 3,836,000  | 5,557,600                                                    | 3,684,800  |
| 5. Air Heater Outlet<br>Flue Gas Temp<br>(°F) (Corrected) | 250                 | 250        | 300                                                          | 300        |
| 6. Air Heater Outlet<br>Dust Loading<br>(lb/hr)           | 33,532              | 21,670     | 30,734                                                       | 19,697     |
| 7. Air Heater Outlet<br>Flue Gas Pressure<br>(in. water)  | -15.25              | -8.28      | -14.82                                                       | -8.16      |
| 8. Minimum Boiler Load                                    | 40%                 |            | 40%                                                          |            |

B. Coal

|                                     | <u>Wyoming Coal</u> | <u>Oklahoma Coal</u> |
|-------------------------------------|---------------------|----------------------|
| 1. <u>Proximate Analysis, Wt. %</u> |                     |                      |
| Moisture                            | 31                  | 7.7                  |
| Ash                                 | 6.01 (6.0-12.0)     | 8.3 (5.0-15.0)       |
| Volatile Matter                     | 30.05               | 35.5                 |
| Fixed Carbon                        | 32.94               | 48.5                 |
| Sulfur                              | 0.40 (0.2-1.0)      | 2.69 (0.8-4.0)       |
| Btu per Pound                       | 8,055               | 12,000               |

TABLE 1 (Cont'd)

B. Coal (Continued)

|                                                       | <u>Wyoming Coal</u> | <u>Oklahoma Coal</u> |
|-------------------------------------------------------|---------------------|----------------------|
| 2. <u>Ultimate Analysis, Wt. %</u>                    |                     |                      |
| Moisture                                              | 31                  | 7.7                  |
| Carbon                                                | 46.81               | 69.5                 |
| Hydrogen                                              | 3.25                | 5.0                  |
| Nitrogen                                              | 0.65                | 1.4                  |
| Chlorine                                              | .01                 | .14                  |
| Sulfur                                                | 0.40 (0.2-1.0)      | 2.69 (0.8-4.0)       |
| Ash                                                   | 6.01 (6.0-12.0)     | 8.3 (5.0-15.0)       |
| Oxygen                                                | 11.86               | 5.27                 |
| 3. <u>Mineral Analysis of Ash, Wt. %</u>              |                     |                      |
| P <sub>2</sub> O <sub>5</sub>                         | 0.52                | -                    |
| SiO <sub>2</sub>                                      | 34.93               | 40.5                 |
| Fe <sub>2</sub> O <sub>3</sub>                        | 6.02                | 23.5                 |
| Al <sub>2</sub> O <sub>3</sub>                        | 16.64               | 23.5                 |
| TiO <sub>2</sub>                                      | 1.02                | 1.0                  |
| CaO                                                   | 20.38               | 4.6                  |
| MgO                                                   | 4.62                | 1.0                  |
| SO <sub>3</sub>                                       | 14.65               | -                    |
| K <sub>2</sub> O                                      | 0.51                | 1.75                 |
| Na <sub>2</sub> O                                     | 1.11                | .35                  |
| 4. <u>Alkalies as Na<sub>2</sub>O, Wt. % (O.C.B.)</u> | 0.13                | .04                  |
| 5. <u>Fusion Temperatures of Ash, °F</u>              |                     |                      |
| <u>Reducing</u>                                       |                     |                      |
| ID                                                    | 2147                |                      |
| H=W                                                   | 2180                |                      |
| H=W/2                                                 | 2192                |                      |
| Fluid                                                 | 2215                |                      |
| <u>Oxidizing</u>                                      |                     |                      |
| ID                                                    | 2212                |                      |
| H=W                                                   | 2234                |                      |
| H=W/2                                                 | 2249                |                      |
| Fluid                                                 | 2279                |                      |
| <u>T<sub>250</sub>, °F</u>                            |                     | 2325                 |

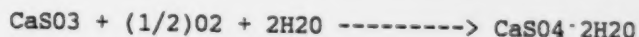
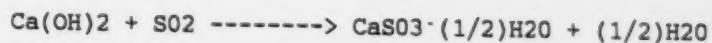
TABLE 1 (Cont'd)

|                                        | <u>Wyoming Coal</u>                                                | <u>Oklahoma Coal</u> |                               |             |             |             |
|----------------------------------------|--------------------------------------------------------------------|----------------------|-------------------------------|-------------|-------------|-------------|
| 6. <u>Equilibrated Moisture, Wt. %</u> | 29.64                                                              | 3.0                  |                               |             |             |             |
| 7. <u>Silica Value</u>                 | 0.53                                                               |                      |                               |             |             |             |
| 8. <u>Coal Size to Feeder</u>          | 1-1/4" x 0"                                                        | 1-1/4" x 0"          |                               |             |             |             |
| 9. <u>Hardgrove Grindability</u>       | 52.0                                                               | 55                   |                               |             |             |             |
| C. <u>Lime</u>                         |                                                                    |                      |                               |             |             |             |
| Type:                                  | Pebble lime (AWWA B202)                                            |                      |                               |             |             |             |
| Size:                                  | 3/4" or less                                                       |                      |                               |             |             |             |
| Available CaO:                         | 85%                                                                |                      |                               |             |             |             |
| Slaking rate:                          | 40°C rise in 3 minutes and slaking reaction complete in 10 minutes |                      |                               |             |             |             |
| Solids & residue:                      | 15% maximum                                                        |                      |                               |             |             |             |
| Density:                               |                                                                    |                      |                               |             |             |             |
| Volumetric -                           | 55 PCF                                                             |                      |                               |             |             |             |
| Structural -                           | 110 PCF                                                            |                      |                               |             |             |             |
| D. <u>Makeup Water</u>                 |                                                                    |                      |                               |             |             |             |
|                                        | <u>Service Water</u>                                               |                      | <u>Cooling Tower Blowdown</u> |             |             |             |
|                                        | <u>Min.</u>                                                        | <u>Avg.</u>          | <u>Max.</u>                   | <u>Min.</u> | <u>Avg.</u> | <u>Max.</u> |
| Total Dissolved Solids                 | -                                                                  | 84                   | -                             | 537         | 831         | 1382        |
| Total Suspended Solids                 | -                                                                  | 9                    | -                             | 10          | 15          | 31          |
| Calcium (as CaCO3)                     | -                                                                  | 90                   | -                             | 140         | 370         | 650         |
| Magnesium (as CaCO3)                   | -                                                                  | 28                   | -                             | 50          | 140         | 290         |
| Hardness (as CaCO3)                    | -                                                                  | 118                  | -                             | 206         | 510         | 1400        |
| Alkalinity                             | -                                                                  | 88                   | -                             | 10          | 112         | 139         |
| Chlorides                              | -                                                                  | 15                   | -                             | 33          | 71          | 94          |
| Sulfates                               | -                                                                  | 35.4                 | -                             | 613         | 717         | 820         |
| pH                                     | -                                                                  | 7.0-7.6              | -                             | 7.0         | 7.0-7.6     | 8.7         |
| Temperature (F)                        | 42                                                                 | 58                   | 84                            | 68          | 81.4        | 94          |
| E. <u>Site Conditions</u>              |                                                                    |                      |                               |             |             |             |
| 1. Altitude:                           | 624' above MSL                                                     |                      |                               |             |             |             |
| 2. Temperature:                        | -5 to 110°F                                                        |                      |                               |             |             |             |
| 3. Relative Humidity:                  | 0-100%                                                             |                      |                               |             |             |             |
| F. <u>Emission Requirements</u>        |                                                                    |                      |                               |             |             |             |
| SO <sub>2</sub>                        | 85% Removal or .6 lb/mm BTU                                        |                      |                               |             |             |             |
| Particulate                            | .03 lb/mm BTU                                                      |                      |                               |             |             |             |



top of each reactor. Atomizers at the top of each reactor spray liquid reagent into the hot flue gas. The reagent dries and, at the same time, chemically reacts with the sulfur dioxide (SO<sub>2</sub>) in the flue gas. Through this reaction, the sulfur dioxide is removed from the flue gas. The rest of the dry product and fly ash is removed in the electrostatic precipitators. The dry product and fly ash is then transported either to the recycle system or the fly ash storage silos for disposal.

The process chemistry associated with the removal of sulfur dioxide from the flue gas is a simple acid/base absorption reaction. When the slurry mixture of lime, fly ash and water is finely atomized in the spray dryer, it intimately mixes with the flue gas. A chemical reaction occurs between the sulfur dioxide and calcium hydroxide according to the following:



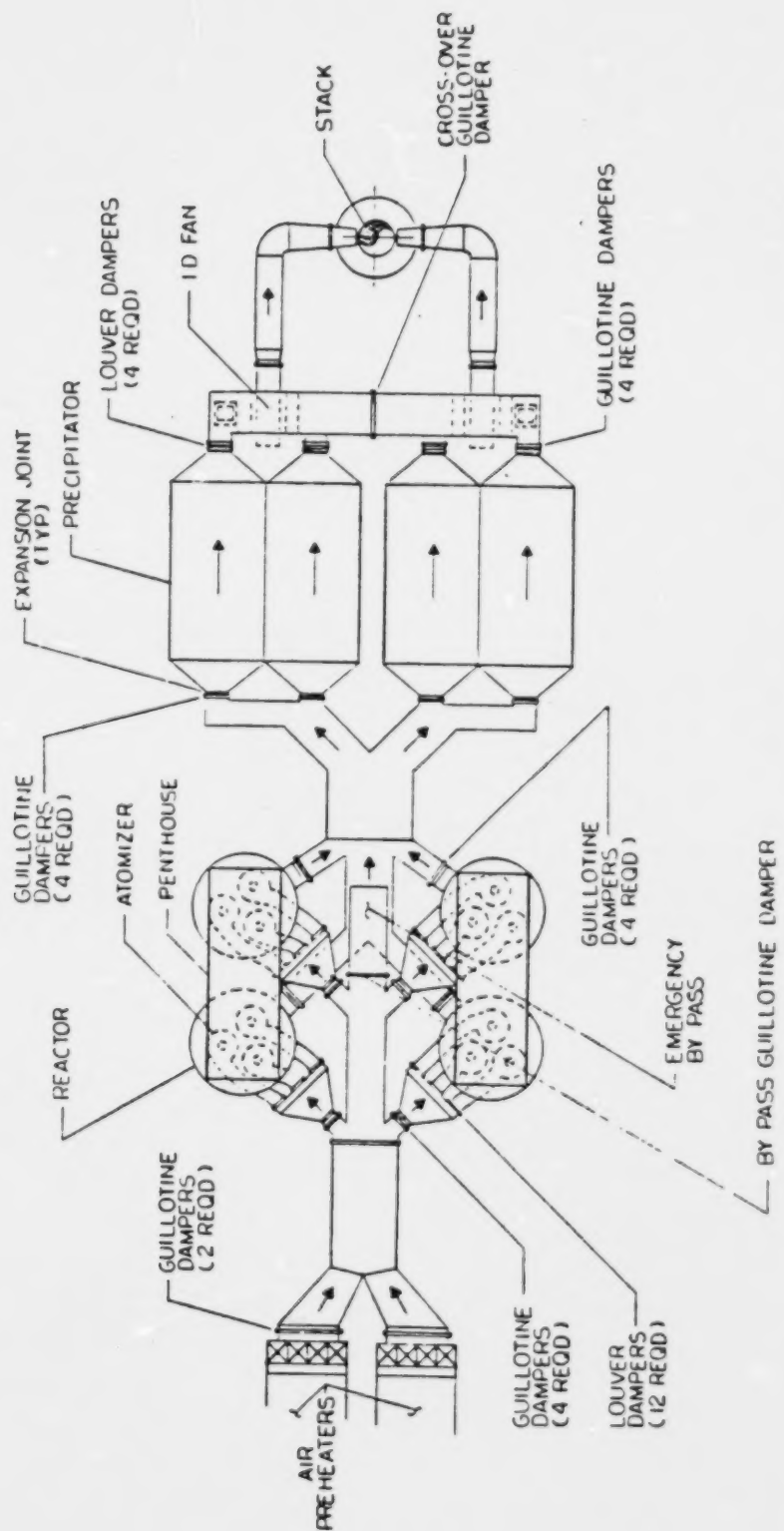
The calcium hydroxide is produced from pebble lime or calcium oxide which is slaked with an excess of water to product a milk-of-lime, or lime slurry. The chemical reaction for this is stated as follows:



The absorption chemistry is affected by several parameters including flue gas temperature, SO<sub>2</sub> concentration in the flue gas, atomized slurry droplet size, distribution of flue gas around each atomizer and the density of the reactor feed slurry.

A schematic diagram of the flue gas flow path is illustrated in Figure 2. Four spray dryer reactors are included in the system, providing thirty-three per cent spare capacity. Each reactor is fifty-two (52) feet in diameter and thirty-five (35) feed straight side providing twelve seconds flue gas residence time. Flue gas leaves the economizer outlet, and enters the top of each reactor. At the top of the reactor, the flue gas is equally distributed to each of three inlet dispersers. The flue gas velocity accelerates as it passes through the disperser. At the discharge of the disperser, where finely atomized droplets of slurry are introduced, the gas velocity is abruptly reduced. This causes a very high degree of turbulence which assures that the droplets become intimately mixed with the flue gas. Individual reactors are capable of providing optimum atomization and gas/slurry mixing over a two to one (2:1) turn-down range.

One rotary atomizer is installed in each of the three dispersers of each reactor. Two additional spare atomizers are provided. The rotary atomizers are a Flakt design. The atomizers are equipped with 300 Hp motors (1800 rpm), mechanical gearbox speed increasers and frequency inverters for speed adjustment. Each atomizer is equipped with a integrated oil lubrication system and is fully instrumented. The design range of the atomizers is 7000-13,000 rpm and 0 to 32,000 lbs/hr slurry flow. The gearbox design is a two stage gear system utilizing helical gears. All high speed bearings are oil filled sleeve bearings which give longer life than ball or roller bearings at high speed. The atomizer disks are 360 mm diameter (O.D.) and are provided with wear resistant ceramic nozzles, bottom and top liners.



FLUE GAS SYSTEM

FIGURE 2

The flue gas leaves the side of the reactor and dust fall-out is removed from the bottom cone discharge. A crusher is provided, at the bottom of the reactor hopper, to handle any large lumps of ash, and is connected directly to a double-dump valve, which serves as the pressure isolation valve. The dust is then conveyed directly to the recycle silo. The rated capacity of the ash conveying equipment is 40,800 lb/hr.

A warm gas by-pass system is provided which can divert reactor inlet flue gas around each reactor and mix it with cooled reactor gas. The system is designed to increase the gas by nine degrees Fahrenheit.

In order to assure that optimum operation of the spray dryer reactors is maintained, and that upset conditions are minimized, the slurry flow to each reactor is individually controlled based on the flue gas temperature exiting each reactor. Gas flow model studies, and field testing were carried out to assure that gas is equally distributed to each of the dispersers of each reactor under all load conditions and combinations of reactors in operation. In addition, the process control system is designed to assure that, correspondingly, equal slurry flow distribution to each of the atomizers is also achieved. This results in a proper matching of the flue gas and slurry flows, thus preventing over or under-spraying which can cause "wet-bottom" in the reactor, or poor absorption of  $\text{SO}_2$ .

The slurry preparation system flow path is illustrated in Figures 3 and 4. The reactor slurry feed system is comprised of the reactor slurry feed tank, reactor feed pumps (rated at 1100 gpm) and reactor head tanks. A mixture of recycle slurry and lime slurry is pumped from the reactor slurry feed tank, in a circulating loop, to all the reactor head tanks. The required slurry is drawn off from the reactor head tanks to each of the atomizers. The remaining slurry mixture overflows from the head tanks and gravity feeds back to the reactor slurry feed tank. The amount of recycle slurry added is directly controlled by the level in the tank. The amount of lime slurry is controlled by the  $\text{SO}_2$  removal performance of the system. Both feed forward and trim-back control logic is utilized to assure optimum lime utilization.

Recycling the reaction products and fly ash helps the drying process and allows for even better utilization of lime as not all of the lime reacts with  $\text{SO}_2$  the first time it enters the reactors. In addition, the fly ash contains some lime which can be used to reduce the amount of fresh lime added. The reactor product that is removed from the bottom of each reactor is conveyed to the recycle storage bin. Particulate collected in the electrostatic precipitator is also diverted to the recycle storage bin as required. The recycle dust is fed from the storage bin through the recycle feeder conveyor in the recycle mix tank where it is combined with water to produce a fixed slurry concentration typically in the range of 30 to 40% solids. Depending on the operating conditions, slurry as high as 50% can be produced. The recycle slurry overflows from the recycle mix tank into a recycle classifier which separates out over-sized solids. The classified recycled slurry then falls directly into the reactor feed tank.

Pebble lime can be delivered by truck or railroad car and is unloaded at the dry lime unloading area remote from the FGD system. The dry lime is conveyed into one of two lime storage silos (each with 1600 tons capacity). The lime in these silos is conveyed to either of two lime day silos located in the lime slurry preparation area. The lime day silos supply lime to either of two ball mill slakers. The ball mill slakers,

FIGURE 3

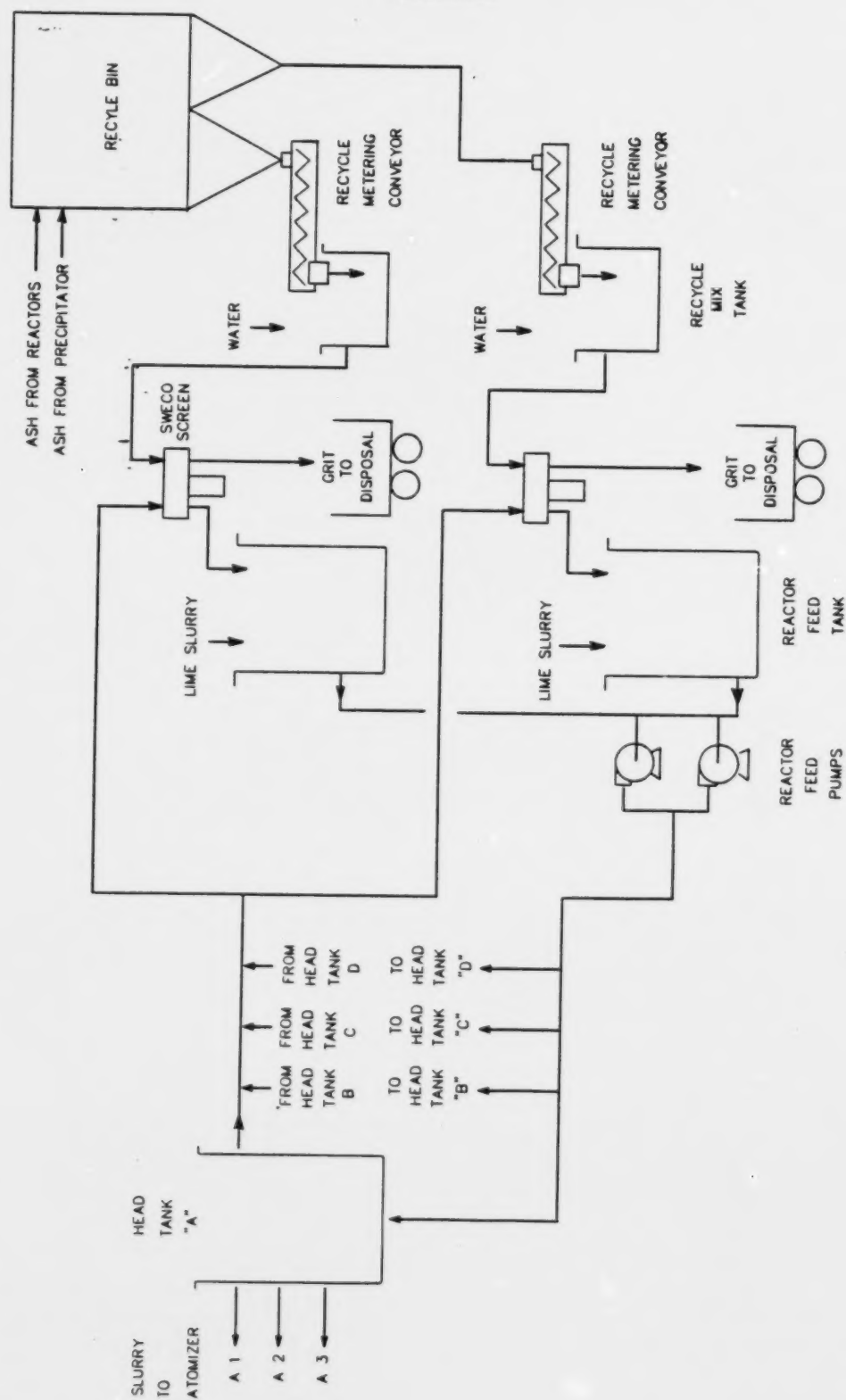
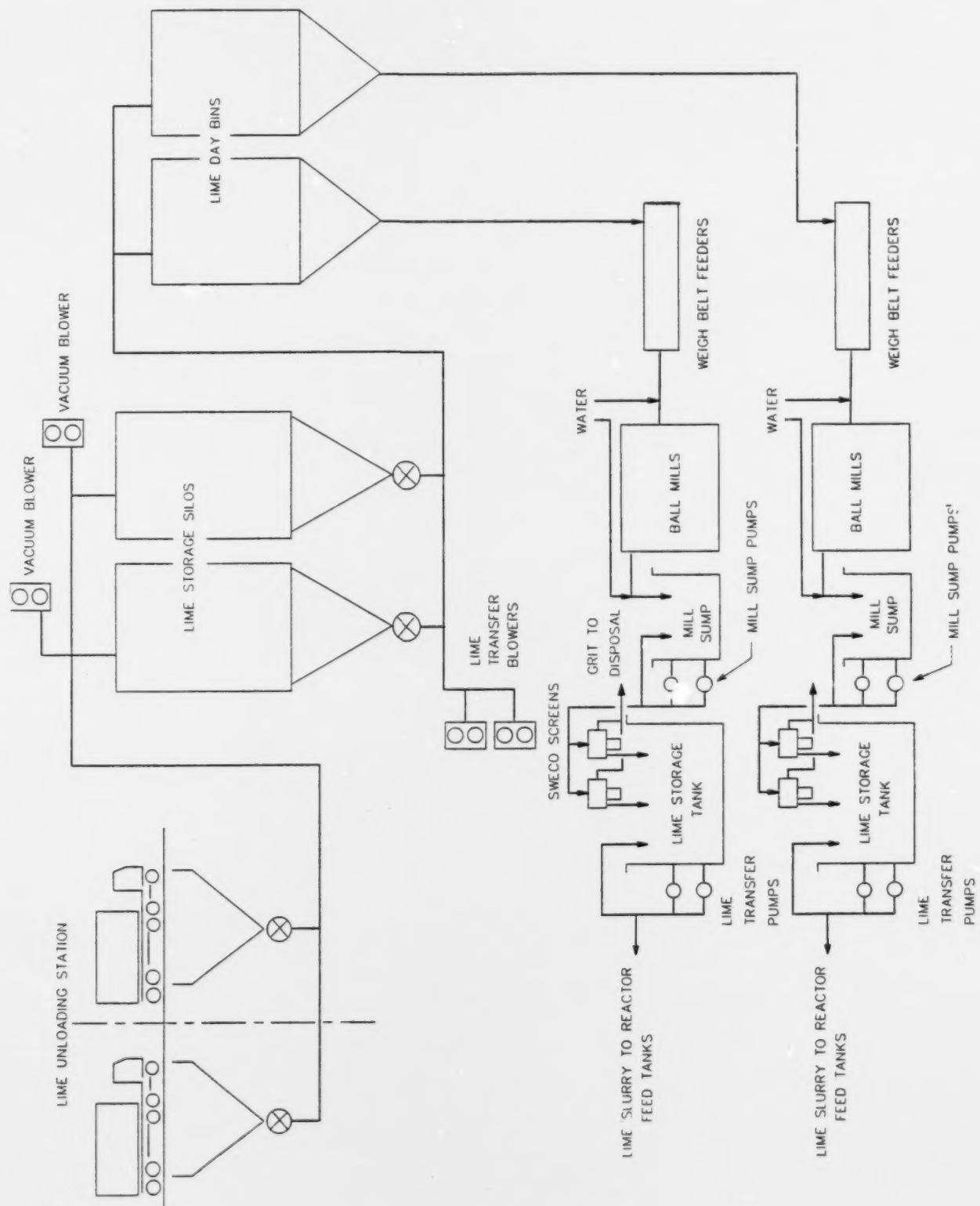




FIGURE 4



supplied by KVS have a rated capacity of 20,000 lbs/hr, each. The operation of the slaking system is in batch mode, based on the level of the lime slurry storage tank. Pebble lime is metered into the ball mills, with a corresponding amount of water. The slaking temperature is maintained at about 170-180 degrees Fahrenheit. The lime slurry exiting from the ball mill is diluted to about 20% solids and stored in the lime storage tank.

The complete slurry preparation and distribution (lime and recycle), is 100% spared with a high degree of crossover possibility.

The particulate control system is comprised of two dual chamber rigid frame electrostatic precipitators operating in parallel. Each chamber has five electrical and mechanical fields in series and two cells in width. Each bus section is independently energized. Rapping systems for the gas distribution screen, collecting electrodes and discharge electrodes are of the internal tumbling hammer design.

The ash handling system includes mechanical conveying equipment (drag chain) and pneumatic conveying. Drag chain conveyors are used to convey the reactor product from the reactor to the recycle silo. All dust collected in the electrostatic precipitator is conveyed by a UCC supplied pressure pneumatic conveying system. The dust is conveyed either to the recycle silo according to level in the silo. Otherwise, the dust is transmitted to the waste ash silo. The ash conveying system is rated at 80 tons per hour.

Waste ash is stored separately from the flyash collected from Unit #1, and is trucked to a nearby ash pond.

The sizing of the various APC System components is described in Table 2.

#### OPERATING EXPERIENCE

##### PRECIPITATOR

The precipitator was sized based on collecting fly ash from untreated flue gas. This results in an over-sized unit for scrubbed flue gas. There has been no problem meeting opacity requirements. At times, the system has maintained compliance with only 20 of 40 bus sections energized. Normal operation is with all fields energized at reduced power under control of the energy management system.

The only maintenance issue to date is corrosion of the insulator compartments and access doors. This has been controlled by increasing the temperature of the insulator heaters. An ultimate solution for future systems would be to provide heated purge air for the insulator compartments and to epoxy coat the access doors.

##### ASH HANDLING AND STORAGE

The ash is transported by a UCC dilute phase pressure conveying system. During the early phases of system operation performance tests revealed insufficient conveying capacity. After investigation by GRDA, Flakt, and UCC personnel the cause was isolated in two areas of the system:

1. The vent valves and lines between the transporters and the

TABLE 2

GRDA APC SYSTEM  
EQUIPMENT SIZING

|                            |                                     |
|----------------------------|-------------------------------------|
| Lime Unloading System      | 250 tons per day<br>(12 truckloads) |
| Lime Storage Silos         | 2 @ 1600 tons                       |
| Lime Transfer System       | 2 @ 12.5 TPH                        |
| Lime Day Bins              | 2 @ 100 tons                        |
| Ball Mill Slakers          | 2 @ 10 TPH                          |
| Mill Sump Pumps            | 4 @ 200 gpm                         |
| Ball Mill Classifiers      | 4 @ 60" diameter                    |
| Lime Slurry Storage Tanks  | 2 @ 12,000 gal                      |
| Lime Slurry Transfer Pumps | 4 @ 250 gpm                         |
| Recycle Bin                | 300 tons                            |
| Recycle Feeder Conveyors   | 2 @ 100 TPH                         |
| Recycle Mix Tanks          | 2 @ 6000 gal.                       |
| Recycle Slurry Classifiers | 2 @ 60" diameter                    |
| Reactor Feed Tanks         | 2 @ 4000 gallons                    |
| Reactor Feed Pumps         | 2 @ 1100 gpm                        |
| Reactors                   | 4 @ 52' diameter                    |
| Atomizers                  | 14 @ 300 Hp                         |
| Ash Conveying System       | 2 @ 50 TPH                          |
| Ash Disposal Silos         | 2 @ 1000 tons                       |

## PRECIPITATOR SIZING

|                                                      |            |
|------------------------------------------------------|------------|
| Number of Precipitators .....                        | 2          |
| Chambers per Precipitator .....                      | 2          |
| Mechanical Fields in Series .....                    | 5          |
| Electrical Fields in Series .....                    | 5          |
| Cells per Chamber .....                              | 2          |
| Collector Plate Height .....                         | 45.93 feet |
| Treatment Length .....                               | 61.52 feet |
| Transformer Rectifier Sets<br>per Precipitator ..... | 20         |

hopper were plugged. This resulted in slow and incomplete filling of the transporter, greatly reducing its capacity. The pluggage was caused by deposition of lime and sulfate reaction products on the walls of the pipe and valves. This was accelerated by condensation on these unheated, uninsulated areas. The was solved by replacing the iron pipe and plug valves with rubber hose and pinch valves. The rubber provides enough insulation to reduce the condensation problem, and, because it flexes with each transport cycle, is self cleaning. Pluggage of these lines has not occurred since replacing them with hose.

2. About 30% of the fluidizing stones in the hoppers and transporters were plugged or leaking. This also contributed to slow filling and emptying of the ash transporters. These stones were replaced. A small percentage continue to plug and require replacement as a part of regular maintenance.

After correcting these two areas, the pressure settings and sequencing of the transport system were fine tuned. The ash handling system has been nearly trouble free for a year now and regularly operates at double its rated capacity.

The ash silos are flat bottom with fluidizing air and dry unloading system. There were initially problems with tramp material plugging the unloading chutes and with water leaks into the silo. These were corrected. There has been no problem with ash storage or unloading since that time.

#### LIME UNLOADING AND TRANSFER

The Ducon lime unloading and transfer system consists of a below grade hopper that receives lime from dump trucks or rail cars, a dilute phase vacuum transport system from the hopper to the two, 1600 ton storage silos, and a second dilute phase pressure transport system from the storage silos to the day bins above the slakers. This system initially operated slightly below its rated capacity when transporting from the silos to the day bins. Increasing the motor size on the conveying air blower allowed transferring lime at rated capacity. This increased air flow overloaded the exhaust fan and bin vent on the day bin. This was corrected by connecting the two day bin vents in parallel, thus doubling the filter area available. The exhaust fan speed was also increased.

The lime unloading and conveying system has performed well after the start-up adjustments were completed. It is capable of unloading 12 truckloads per day of pebble lime in any weather, fully controls fugitive dust, and reliably maintains the lime supply to the slakers.

#### LIME SLAKING AND SLURRY HANDLING

The KVS lime slaking system consists of weigh feeders, ball mill slakers, and grit removal screens. Lime transfer pumps and slurry storage tanks transport and store the resulting slurry product.

The slaking system has required a considerable amount of fine tuning to reliably produce good quality lime slurry. Areas addressed include:

1. Plugging in the lime feed chute to the mill. This was a result of the vent system at the mill discharge not generating enough draft to keep steam out of the feed chute. Closing up the many air leaks in the



system improved the ventilation enough to minimize this pluggage.

2. Slurry leaks at the mill inlet seal. Accurate adjustment of the seal and repacking with wear resistant Kevlar packing has controlled this leakage. Also, reduction of outlet trommel screen pluggage (described below) has helped maintain a lower slurry level in the mill and reduced seal leakage.

3. Pluggage of the mill outlet trommel screen. During initial operation of the ball mills GRDA found that operating at the design slurry solids content and temperature in the mill resulted in a viscous slurry that would not pass the trommel screen. Flakt added a row of small high pressure water sprays to continuously wash the trommel screen. This provided enough energy to force the slurry thru the screen without significant dilution. The mills now operate at a slaking temperature of 180°F and maintain a final lime slurry concentration of 20 to 25% after all dilution and screening.

4. The vibrating grit removal screens provided by KVS were a constant maintenance area. They were an inclined, rectangular type, manufactured by FMC, that used vibration along the long axis of the screen to convey the grit to the discharge. These screens plugged easily, were difficult to clean, difficult to replace the screen cloth, allows slurry to flow out the grit discharge, and had a splash containment system that rapidly wore holes in the screens. They were replaced by SWECO Vibro-Energy Separators and the problems were eliminated.

5. A large amount of grit was allowed to pass thru the FMC screens and settle in the lime storage tanks. This eventually plugged the pump suction lines. After replacing the screens and cleaning the tanks, this settling was eliminated.

6. The Philadelphia Mixer motors and gear drives provided for the lime slurry storage tanks were too small to drive the agitator provided without tripping the motor. These were replaced with larger sized units and have performed well since installation.

The ball mill slaker has proven capable of reliably processing large tonnages of lime to produce a high quality lime slurry. Lab tests has shown that the slurry produced is highly available with fine particle size and low settling rate. The mill is able to reduce nearly all the grit to below 40 mesh in size on the first pass. The amount collected on the screens is so small (less than 1% of the total lime feed) that it is simply collected in a dumpster and hauled away rather than being recycled to the mill. The lime pumps, lines, valves, and tanks have had no problem with scaling or wear. The original rubber liner is still in the ball mill and the ball charge has been replaced about one per year.

#### RECYCLE AND FEED SLURRY

Dry product from the reactors and precipitator is conveyed to the recycle bin by drag chain and pneumatic conveyors respectively. It is metered out of the bin by a variable speed chain conveyor to a mix tank. The recycle slurry is passed thru a SWECO screen to remove oversized material. This system required several modifications to achieve reliable performance.

1. The original Philadelphia Mixer agitator installed was too small

to adequately incorporate the light, fluffy, dry recycle into the viscous slurry. It was replaced with one more than three times the original horsepower. This unit has done an outstanding job in a very difficult application. The tank and mixer successfully combine over 100 TPH of dry recycle with 350 GPM of water to produce a slurry with over 50% solids.

2. The recycle product from the reactors contains a significant amount of oversized (greater than 10 mesh) material. This increases the amount of reject material from the screen and increases the wear on all system components. The plant operators have eliminated this material by opening the poke holes on the bottom of the reactor. The air pulled in thru the bottom of the hopper causes all the fine ash to be carried over to the precipitator. The remaining large material is less than one percent of the total dust in the system. This is periodically disposed of with the bottom ash and not recycled.

3. The variable speed conveyor used to meter the dust into the mix tank is not a positive displacement flow control device. At times, the dust becomes very fluid and will flood thru the conveyor. An automatic shut off valve has been installed on the silo as a temporary fix. GRDA is in the process of installing a variable speed rotary valve on the silo to provide positive control of the ash flow rate. The existing conveyor will be replaced by an air slide to simply move the dust from the silo to the tank.

4. The recycle silo has a cone bottom and originally had no other flow inducing devices. Occasionally the silo would plug. Flakt added fluidizing pads to the cone to improve the uniformity of flow.

Problems that did not occur and are notable by their absence are wear, plugging, and scaling of pipes, pumps, and flow control valves. These have been widely reported at other FGD systems. We credit proper design of the slurry system with prevention of these problems. Pipeline velocities were carefully maintained in the range of 5 to 10 FPS. The flow control valves were sized and located so that the pressure drop across the valve is low and the valve normally operates 50 to 100% open. The rubber lined slurry pumps were conservatively sized and operate at only 1200 RPM. They use expeller vanes and a water flushed packed seal on the shaft. To avoid pluggage, the circulating slurry lines were designed to maintain a substantial return flow from the head tanks even when all the atomizers are operating at full load.

#### ATOMIZERS

The atomizers used at GRDA were developed especially for use in FGD service. There were several operating and maintenance issues to be overcome in this application. These were as follows:

1. The piping used in the lubrication system was poorly assembled and leaked at many points. All of the lube oil piping was replaced by qualified workmen under close supervision. This corrected the leaks.

2. After about 6 months of operation three of the atomizers experienced high speed bearing failures. This was primarily caused by contaminated oil. Spot checks of other atomizers revealed dirty oil in units that had not failed. GRDA and Flakt agreed that in order to prevent further failures, all the units should be taken out of service and cleaned. This was done and all new bearings were installed. Additional

operating procedures and automatic alarms were instituted to maintain oil cleanliness and to prevent reoccurrence. No further bearing failures have occurred in 20 months of continuous operation. GRDA has a scheduled inspection program for individual atomizers. The results of this program show that oil cleanliness and bearing life are substantially improved.

3. Atomizer disk wear rates are influenced by recycle ash composition and ash quantity. The Powder River coal used at GRDA has a very abrasive ash. The GRDA system utilizes a very high recycle ash quantity to reduce lime costs. For the purpose of economic use of lime, GRDA chose to operate with double the design recycle rate, and accepted the degradation in the disk wear rate. These two factors require regular maintenance of the atomizer nozzles and wear plates.

Because of the improvements, more recent installations using this machine have in fact not exhibited any of the problems experienced by the GRDA units.

#### REACTORS

Three reactors are required to treat the design gas flow from the boiler. A full spare was provided. During periods when the boiler is operating above design load, all four reactors are placed on line. Each reactor is equipped with three gas dispersers and three atomizers.

The reactors have required a few modifications to optimize operation. Each gas disperser required the addition of a vane to eliminate an eddy current in the gas stream around the atomizer. This eddy current caused an unacceptable lime deposit on the disperser before it was corrected.

Process upsets (over-spraying) have caused occasional wall deposits which later fall into the hopper. An average of 10 hours per month are required to manually clear large pieces. Small chunks less than 1 foot in diameter are ground up by the lump crusher at the hopper outlet without interrupting the operation of the reactor.

The basic reactor sizing of 12 seconds retention time has proven adequate for proper drying and reaction. Flow balancing between the three gas dispersers and atomizers was given careful attention during the design phase of the project, was confirmed by a gas flow model study, and has not been a problem in operation.

#### DUCTWORK AND DAMPERS

The ducts and dampers have been largely trouble free. A gas flow model study predicted the location of turning vanes for proper gas distribution and elimination of ash deposits. Field tests of gas flow confirmed the results of the model. The DDI dampers have performed well and have not been subject to undue wear or corrosion. The ductwork has shown localized corrosion around the expansion joints. This is probably caused by the cooling effect of the exposed joint mounting flanges. These areas are being checked on a regular basis, but do not yet require any repairs.

#### CONTROL SYSTEM

The FGD Control System is a split architecture design using a Honeywell TDC 3000 for analog control and a 4500C for supervisory control.



Digital logic and I/O is primarily handled by an Allen Bradley PLC-3. The Honeywell and the Allen Bradley coordinate their I/O data and control functions over a data highway communications port. Operator interface to the system is by either colorgraphic CRT or conventional panelboard switches and hand/off/auto stations. Total I/O count is approximately 200 analog and 1000 digital. The system has generally performed well. The operators have developed a preference for the CRT interface rather than the conventional panelboard. The main problem reported to date have been hard disk drive and printer failures.

#### STACK ANALYZERS

The Emissions Monitoring System was supplied by Thermo Electron Corporation. Complete monitoring and compliance reporting for SO<sub>2</sub>, NO<sub>x</sub>, Opacity, and CO<sub>2</sub> is performed in the stack. System inlet concentration of SO<sub>2</sub> and CO<sub>2</sub> is measured and used to calculate % removal across the system. The inlet analyzer, a Model 200 Extractive type, has been more reliable than the outlet analyzer, a Model 100 in situ type. Flue gas dewpoint is measured at the inlet to the scrubber by an on-line analyzer. This analyzer has also performed very reliably.

#### PERFORMANCE RESULTS

As part of the contractual agreement between GRDA and Flakt, the FGD system was to meet certain operating and performance guarantees. In order to obtain necessary data to prove that the AQCS system met its performance guarantees, GRDA requested Benham-Holway to prepare and execute a mutually agreeable test protocol. The tests were carried out in March of 1987. The results of the tests are shown in Table 3 and 4 together with the guarantee values.

The Contract required that SO<sub>2</sub> and particulate efficiencies be met on a make right basis. GRDA also incorporated liquidated damages with debit/credit netting for power and consumables into the Contract.

The overall results of the AQCS system with respect to the assessment of performance shows that the AQCS system passed the performance test in all respects requiring no remedial corrective work and resulting in a net test credit for consumables to Flakt of \$39,996,550. Over an anticipated plant life of thirty (30) years, this amounts to a projected annual operating savings of approximately \$1.33 million year. These cost savings are detailed in Table 5. Although of no monetary value to Flakt, it shows that the FGD system supplied by Flakt does perform the emissions controls function for which it was designed. Furthermore, it will do so at a substantially lower lifetime cost to the Authority than was originally anticipated.

#### DISCLAIMER

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.



TABLE 3  
OVERALL AQCS TEST RESULTS  
VERSUS  
CONTRACT GUARANTEES

|                                         | <u>Test</u>   | <u>Contract</u> |
|-----------------------------------------|---------------|-----------------|
| Power Consumption (kW)                  | <u>3362.8</u> | <u>5042.0+</u>  |
| Pebble Lime Consumption (lbs/hr)        | <u>11932</u>  | <u>19431+</u>   |
| Pressure Drop (in H <sub>2</sub> O)     | <u>9.8*</u>   | <u>11.5+</u>    |
| SO <sub>2</sub> Removal Efficiency (%)  | <u>87.8</u>   | <u>85.0</u>     |
| Recycle Capability (lbs/hr)             | <u>*</u>      | <u>70,000</u>   |
| Average Opacity (%)                     | <u>5.53</u>   | <u>20</u>       |
| Chimney Particulate Level (lbs/MBTU)    | <u>*</u>      | <u>.02785</u>   |
| Outlet SO <sub>2</sub> Level (lbs/MBTU) | <u>.337</u>   | <u>.600</u>     |
| Stoichiometric Ratio                    | <u>1.17</u>   | <u>1.57+</u>    |
| Service Water Consumption (GPM)         | <u>136.9</u>  | <u>141.1+</u>   |
| Clg. Twr. Blowdown Consumption (GPM)    | <u>164.7</u>  | <u>219.9+</u>   |
| Approach Temperature (°F)               | <u>26.2</u>   | <u>20</u>       |

\*Not measured during the performance tests. Accepted based upon AQC system performance during the compliance tests.

+Corrected contract values.

**TABLE 4**  
**ELECTROSTATIC PRECIPITATOR PERFORMANCE**

|                                                         | <u>Test No.</u><br><u>1</u> | <u>Test No.</u><br><u>2</u> | <u>Test No.</u><br><u>3</u> |
|---------------------------------------------------------|-----------------------------|-----------------------------|-----------------------------|
| Gas Flow Rate (ACFM)                                    | 1,780,000                   | 1,828,000                   | 1,829,000                   |
| Inlet Gas Temperature (°F)                              | 159                         | 156                         | 156                         |
| Inlet Particulate<br>Concentration (gr/DSCF)            | 10.57                       | 10.45                       | 11.66                       |
| Outlet Particulate<br>Concentration (gr/DSCF)           | 0.0040                      | 0.0047                      | 0.0038                      |
| (lb/MBTU) *                                             | 0.0078                      | 0.0098                      | 0.0073                      |
| Precipitator Removal<br>Efficiency (%)                  | 99.96                       | 99.96                       | 99.97                       |
| Opacity (%)                                             | 1.5                         | 1.5                         | 1.5                         |
| Transformer Rectifier **<br>Power Consumption (KWHR/HR) | 505                         | 509                         | 513                         |
| Specific Power Consumption<br>(Watts/1000 ACFM)         | 284                         | 278                         | 281                         |

\* By the Fd Factor Method

\*\* Semipulse /Energy Management

TABLE 5

## OVERALL AQCS TESTING ASSESSMENTS

|                                         | Performance<br>Differential | Contractual<br>Performance<br>Assessments | Credit       | Debit |
|-----------------------------------------|-----------------------------|-------------------------------------------|--------------|-------|
| Power Consumption (kW)                  | 1679.2                      | \$6,000/kW                                | \$10,075,200 |       |
| Pebble Lime Consumption (lbs/Hr)        | 7499                        | 3,650/lb/hr                               | 27,371,350   |       |
| Pressure Drop (in H <sub>2</sub> O)     | 1.7                         | 1,500,000/in H <sub>2</sub> O             | 2,550,000    |       |
| SO <sub>2</sub> Removal Efficiency (%)  | 2.8                         | *                                         |              |       |
| Recycle Capability (lbs/Hr)             | --                          | *                                         |              |       |
| Average Opacity (%)                     | 14.47                       | *                                         |              |       |
| Chimney Particulate Level (lbs/MBTU)    | --                          | *                                         |              |       |
| Outlet SO <sub>2</sub> Level (lbs/MBTU) | .263                        | *                                         |              |       |
| Stoichiometric Ratio                    | .40                         | *                                         |              |       |
| Service Water Consumption (GPM)         | 4.2                         | *                                         |              |       |
| Clg. Twr. Blowdown Consumption (GPM)    | 55.2                        | *                                         |              |       |
| Approach Temperature (°F)               | 6.2                         | *                                         |              |       |
|                                         |                             | TOTAL                                     | 39,996,550   |       |

\*No monetary penalty involved. The contract required remedial corrective action by the Contractor to bring the systems into compliance should they not meet contract requirements.

\*\*A positive performance differential indicates performance in excess of contract requirements.

# EVALUATION OF A COMBINED $\text{Ca(OH)}_2/\text{NH}_3$ SPRAY DRYER SYSTEM FOR $\text{SO}_2$ REMOVAL ON HIGH SULFUR COAL

by: Wayne T. Davis, Gregory D. Reed, Arijit Pakrasi  
Civil Engineering Department  
the University of Tennessee  
Knoxville, TN 37996

and

Timothy C. Keener  
Civil and Environmental Engineering Department  
University of Cincinnati  
Cincinnati, OH 45221

## ABSTRACT

The objective of this research was to develop a more cost effective alternative to  $\text{Ca(OH)}_2$ -based spray dryer technology for removal of sulfur dioxide ( $\text{SO}_2$ ) from high sulfur coal. The alternative proposed was that of a baseline  $\text{Ca(OH)}_2$  system with an ammonia ( $\text{NH}_3$ ) additive injection system. This system utilized a standard spray dryer system for the initial  $\text{SO}_2$  removal, and supplemented the removal with gaseous  $\text{NH}_3$ . This mode of operation allowed use of  $\text{Ca(OH)}_2$  up to a stoichiometric ratio of 0.6 - 0.7 at which point increases in stoichiometric ratio have been shown to result in decreased utilization. The  $\text{NH}_3$  was injected as a second additive, also reacting with  $\text{SO}_2/\text{SO}_3$  to form ammonium sulfates/sulfites which sublimed to solid particles which were then removed in a fabric filter collector to obtain the overall  $\text{SO}_2$  removal efficiency. An additional objective was to demonstrate that the combined  $\text{Ca(OH)}_2/\text{NH}_3$  system could achieve 90%  $\text{SO}_2$  removal on high sulfur coal ( $\text{SO}_2$  concentrations of 2000-3000 ppm), a condition which cannot be achieved with a system using only  $\text{Ca(OH)}_2$  in a spray dryer/fabric filter.

The objective was accomplished by conducting tests on a 1000 ACFM spray dryer/fabric filter collector to 1) evaluate the stoichiometric requirements of the combined  $\text{Ca(OH)}_2/\text{NH}_3$  system, 2) demonstrate its effectiveness on higher sulfur coals (up to 3000 ppm), 3) study the waste characteristics of the combined  $\text{Ca}/\text{NH}_3$ -based waste, and 4) evaluate the potential of  $\text{NH}_3$  regeneration by physical/chemical treatment of the waste.



## BACKGROUND

### Ca(OH)<sub>2</sub> STUDIES

Numerous studies have been conducted at both full scale and at the pilot scale demonstrating the relationship between the SO<sub>2</sub> removal efficiency and the operating parameters associated with spray dryer/fabric filter systems(1,2,3). In general, these studies have shown that the efficiency increases with increased stoichiometric ratio and with nearer approaches to saturation at the spray dryer outlet. In addition, the fabric filter serves as a dry reactor due to the increased residence time of the dried solids on the filter bags. For purposes of background development, this paper will use data generated at the 1000 ACFM pilot spray dryer/fabric filter facility located at the University of Tennessee (to be described in a later section).

Typical spray dryer/fabric filter performance data for SO<sub>2</sub> removal efficiency versus stoichiometric ratio (SR) of Ca(OH)<sub>2</sub> are shown in Figure 1 for concentrations of 2790-3100 ppm of SO<sub>2</sub>. The operating conditions of the spray dryer for the data were as follows: an inlet temperature of 300°F, an approach to saturation of 30°F and no recycle. The removal efficiency of the spray dryer was approximately 27% at a SR of 0.45 resulting in a utilization of only 55%. The spray dryer efficiency increased to 48% at a SR of 1.2, however the utilization had decreased to only 40%. The system, consisting of the spray dryer and fabric filter collector, had an overall utilization of 82% and 48% at SRs of .45 and 1.2, respectively. While utilization improved with the system, it is important to note that without recycle the efficiency of the system is far less than the 90+% that is typically required for a high sulfur coal application. Since the slurry concentration at a SR of 1.2 is approaching 30%, it is evident that the system cannot achieve the required efficiency.

### NH<sub>3</sub> STUDIES

In an effort to improve the utilization of the Ca(OH)<sub>2</sub>-based spray dryer, it was hypothesized that if the system was operated at a SR where the utilization was near 100%, and if a suitable additional reactant could be identified to be used in a supplemental mode, then one might be able to achieve required efficiencies with improved utilization. Based on data obtained previously in all-dry gaseous NH<sub>3</sub> injection studies, it was proposed that NH<sub>3</sub> be used. Research conducted in 1973 by Shale(4,5) and in 1976 by the authors(6,7) has shown, under proper conditions of flue gas temperature, that gaseous NH<sub>3</sub> will react with gaseous SO<sub>2</sub> and sublime to a fine particulate solid, resulting in the removal of SO<sub>2</sub>. In those studies it was shown that the NH<sub>3</sub> and SO<sub>2</sub> sublimed to a solid at a temperature of 180-190°F. At flue gas temperatures below 180°F, a fabric filter effectively removed the reacted products. At temperatures above the critical value, a visible plume occurred, consisting of sub-micron particles, presumed to be comprised of ammonium sulfite/sulfate and/or ammonium bisulfite/bisulfates.

As part of the current study, tests were conducted in the 1000 ACFM pilot plant in which the temperature of the system was controlled by water injection (no calcium hydroxide) to achieve a spray dryer outlet temperature of 155°F (a 30°F approach to saturation). The operating conditions were the same as those discussed in Figure 1. Figure 2 shows a graph of SO<sub>2</sub> removal efficiency versus stoichiometric ratio of NH<sub>3</sub>, assuming that two moles of NH<sub>3</sub> were required to react with one mole of SO<sub>2</sub>. Based on this assumption, a system efficiency of 40-50% was achieved at a SR of 0.4-0.5 of NH<sub>3</sub>, indicating a near perfect utilization. This reconfirmed the data reported previously(5,6,7).

## FACILITIES AND TEST PROCEDURES

### DESCRIPTION OF THE PILOT PLANT

The pilot plant consisted of a nominal 1000 ACFM system located on a sidestream of the University of Tennessee Riley stoker-fired boilers. The system shown in Figure 3 consisted of a slaking system, a spray dryer (7 feet in diameter) with a variable speed Stork-Bowen AA-6 Atomizer (17,000 rpm, 6 inch centrifugal atomizer), and a 36 bag fabric filter collector operated at an air/cloth ratio of 2.9/1 or 2.9 fpm. The collector contained 13.6 ounce/yd<sup>2</sup> fiberglass filter bags which were cleaned with a low energy compressed air supply (10-13 psi). For the data collected herein, the collector was operated for 4-6 hours between cleaning in a pressure drop range of 3-6 inches of water.

The system provided for continuous monitoring of the static pressure, temperature, and SO<sub>2</sub> concentrations at the inlet and outlet of the spray dryer and baghouse. Continuously extracted samples of flue gas were transported through heat-traced lines to a TECO Model 900/Model 40 SO<sub>2</sub> analyzer. The approach to saturation was measured at all three ports using a hand-held wetted wick thermometer and a thermocouple.

The concentration of SO<sub>2</sub> was maintained by supplementing the boiler's baseline 400-500 ppm by using SO<sub>2</sub> supplied from liquid storage tanks. Ammonia injection was accomplished by monitoring the ammonia injection rate from liquid ammonia storage tanks. In the testing reported in this study, the ammonia was injected upstream of the spray dryer, but downstream of the SO<sub>2</sub> supplemental injection port and inlet sampling port.

### TEST PROCEDURES

All data reported represent individual tests of 45-60 minutes in duration. A single data set consisted of monitoring disk speed, slurry injection rate, SO<sub>2</sub> concentrations at three ports, temperature and wet-bulb temperature at three ports, orifice plate pressure drop (flow rate), available CaO, static pressure at three ports, and the ammonia flow rate.

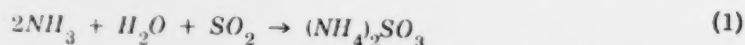
In those tests in which ammonia was injected, the system was operated in an ammonia starved mode at all times that sampling was conducted. This was mandated by the fact that if unreacted ammonia was present, it would pass through the sintered steel filters located at the entrance to the heat trace lines at each sampling port. This would create potential errors in the sampling as reactions would occur in the lines, and would have caused particulate formation in the analyzer. It was possible to achieve an ammonia-starved condition by assuring that two conditions were met: 1) there was always an excess of SO<sub>2</sub> in the exit flue gas, and 2) assuring that there was no visible plume at the exit. In cases where SO<sub>2</sub> was present in the flue gas, the presence of any excess NH<sub>3</sub> resulted in the formation of a visible plume due to the sub-micron particles that were formed at the stack exit as any excess NH<sub>3</sub> reacted with the SO<sub>2</sub>. As a secondary check on the absence of NH<sub>3</sub>, Sensidine tubes were used to sample for NH<sub>3</sub>. While not a very precise technique, the tubes were found to show the presence of NH<sub>3</sub> each time a visible plume was observed.

### STOICHIOMETRIC RATIO

The reaction between Ca(OH)<sub>2</sub> and SO<sub>2</sub> is such that one mole of Ca(OH)<sub>2</sub> is theoretically required to react with 1 mole of SO<sub>2</sub>. The stoichiometric ratio (SR)

is defined as the number of moles of CaO or Ca(OH)<sub>2</sub> actually injected to the theoretical number of moles of CaO required to react with all the SO<sub>2</sub>.

In the case of the NH<sub>3</sub>/SO<sub>2</sub> reaction, the most probable reaction is



On this basis an ammonia stoichiometric ratio (ASR) of 1.0 represented the injection of 2 moles of NH<sub>3</sub> for every 1 mole of SO<sub>2</sub>.

In the case of combined Ca(OH)<sub>2</sub> and NH<sub>3</sub> injection, the combined stoichiometric ratio is calculated as:

$$\text{COMBSR} = \text{SR} + \text{ASR}$$

This calculation method for combined SR permits a comparison of the utilization between single injection systems and combined injection systems as the SR would be calculated on a common basis. For example, a combined stoichiometric ratio (COMBSR) of 1.0 means that there is enough Ca(OH)<sub>2</sub> and NH<sub>3</sub> present to theoretically react with all of the SO<sub>2</sub>.

## RESULTS AND DISCUSSION

### SPRAY DRYER TESTING

The tests were conducted in three phases. In the first phase baseline removal of SO<sub>2</sub> was obtained for both Ca(OH)<sub>2</sub> slurry and NH<sub>3</sub> injection, separately. These were shown in Figures 1 and 2. In the second phase, combined Ca(OH)<sub>2</sub> and NH<sub>3</sub> injection at various ratios were tested to evaluate combined stoichiometric requirements.

In these later tests, the Ca(OH)<sub>2</sub> slurry input was held constant at a preset value and the NH<sub>3</sub> injection rate was slowly increased step wise until either 90% SO<sub>2</sub> removal occurred or a visible white plume was observed at the stack indicating that breakthrough of the NH<sub>3</sub>/SO<sub>2</sub> reaction had occurred. From the results of the above tests, an optimum ratio of Ca(OH)<sub>2</sub> and NH<sub>3</sub> was chosen, which could give 90%+ SO<sub>2</sub> removal at the best sorbent utilization rate. In the third phase, a test was conducted for an extended period at the optimum condition to demonstrate repeatability. In all the above tests, the temperature of the flue gas at the inlet of spray dryer was 300°F ± 10°F and the approach to saturation at the spray dryer outlet was 30°F ± 2°F.

The results of the baseline tests (Figures 1 and 2) showed that 90% SO<sub>2</sub> removal could not be achieved with either a Ca(OH)<sub>2</sub> or an NH<sub>3</sub> system as discussed previously.

It was therefore decided to test the combined system at three different baseline Ca(OH)<sub>2</sub> stoichiometric ratios by injection of NH<sub>3</sub> to achieve the desired 90% SO<sub>2</sub> removal. Figures 4 and 5 show the results of spray dryer and system data for 3000 ppm of SO<sub>2</sub> for the combined system. The solid data points illustrate the baseline Ca(OH)<sub>2</sub> - only data whereas the open circles illustrate the baseline Ca(OH)<sub>2</sub> plus NH<sub>3</sub> data. Figures 6 and 7 show the conditions for the 2000 ppm of SO<sub>2</sub> data.

Figure 5 shows that the combined system can achieve the desired 90% removal with a very good overall sorbent utilization ranging between 95-99% at a baseline



Ca(OH)<sub>2</sub> SR of 0.5-0.7 for 3000 ppm of SO<sub>2</sub> in the flue gas. The overall utilization dropped to 72% at a baseline lime SR of 1.0. Similar decreases in overall utilization occurred for the 2000 ppm data shown in Figure 7.

In phase 1 and phase 2 tests, it was observed that a white visible plume, characteristic of NH<sub>3</sub> breakthrough, occurred when either NH<sub>3</sub> injection was attempted above a 0.45-0.5 SR in the NH<sub>3</sub> only system or when more than 95% system efficiency was attempted in the combined system. The plume disappeared as soon as the NH<sub>3</sub> injection rate was decreased indicating that the system could be operated with 90% efficiency without a visible plume.

The effect of injecting NH<sub>3</sub> just before the baghouse instead of the spray dryer was also investigated in a combined Ca(OH)<sub>2</sub>/NH<sub>3</sub> injection test. The utilization seemed to drop slightly compared to earlier tests. Also NH<sub>3</sub> breakthrough at Port 3 occurred at a slightly lower efficiency. One reason for this could be the lower residence time available for the reaction and sublimation to go to completion. However, additional tests are needed to confirm this. Based on this observation, all tests reported herein were conducted by injecting NH<sub>3</sub> upstream of the spray dryer.

Based on the above results, the extended demonstration test (Phase 3) was conducted at a baseline Ca(OH)<sub>2</sub> SR of 0.7-0.8 with supplemental NH<sub>3</sub> injection to achieve 90% removal efficiency without a plume. This testing was carried out for a 7 hour period at stable conditions during two consecutive days. The results (Figure 8) showed that 90% removal was obtained with an average combined SR of 0.97. The contributions of the spray dryer and fabric filter were 79% and 11%, respectively. The test conditions were 1040-1110 ACFM of flue gas with 2600-3000 ppm inlet SO<sub>2</sub> at 285-310°F and a 30°F ± 2°F approach to saturation. The average overall sorbent utilization was 91.6% and the range was 85.6% to 99% at varying baseline Ca(OH)<sub>2</sub> SRs of 0.7-0.8. The average SO<sub>2</sub> removal efficiency was 90.3%. There was no visible plume under this condition.

One of the primary concerns associated with the supplemental NH<sub>3</sub> injection is the formation of submicron particles that might eventually lead to higher pressure drop across baghouse. Tests showed that the pressure drop rise rate of the combined system was similar to that of the Ca(OH)<sub>2</sub> - system indicating a buffering effect by the larger sized Ca(OH)<sub>2</sub>/SO<sub>2</sub> product. The initial pressure drop in these tests ranged from 3-4 "wg and the rise rate was approximately 0.375 "wg/hr at an air to cloth ratio of 2.9-3.1 cfm/ft<sup>2</sup> and an average dust load of 6.0-6.4 grains/CF in the flue gas. Additional tests are needed to determine the long-term effects of the combined system on the filter bags.

#### CHARACTERIZATION OF WASTE PRODUCTS

The waste collected from the pilot plant during operation was subjected to a variety of tests to determine the physical characteristics and specific chemical component analyses. Particle sizing experiments were also conducted with a Pilot Mark 3 Cascade impactor(8). These data are summarized in Table 1.

These results indicate that the wastes are similar with respect to surface area, density, bulk density, and percent water due to the predominance of the Ca-based waste. Total sulfur analyses were conducted at two different decomposition temperatures (oven temperatures) of 820 and 2600°F. The low sulfur values obtained at lower temperature indicate that the fly ash and Ca(OH)<sub>2</sub> products are more stable than the NH<sub>3</sub> product.



TCLP extraction tests were conducted on the wastes taken from both the spray dryer and the baghouse. The results are shown in Table 2. The values listed are specific for the system and could vary considerably for other coal ashes. The results indicate that concentrations of all the metals except selenium are well below the RCRA maximum concentration limits. The selenium level was highest in the only  $\text{NH}_3$  waste and tended to decrease with increasing calcium concentration. This suggests that a system designed to regenerate  $\text{NH}_3$  from the waste using calcium oxide will have the added advantage of fixing the waste.

### $\text{NH}_3$ REGENERATION

Studies are underway to evaluate the possible regeneration of the  $\text{NH}_3$  using a calcium-based regeneration scheme. The results were not yet available at the time of this writing.

### CONCLUSIONS

1. A novel  $\text{SO}_2$  removal technique using baseline removal with  $\text{Ca}(\text{OH})_2$  slurry and supplemental removal by  $\text{NH}_3$  has been demonstrated in a 1000 ACFM pilot plant with 90% removal efficiency at an average of 91.6% utilization for 2600-3000 ppm of  $\text{SO}_2$  in the flue gas at  $30^\circ \pm 2^\circ\text{F}$  approach to saturation.
2. The molar reaction ratio of  $\text{NH}_3/\text{SO}_2$  at this condition was determined to be 2:1.
3. A visible plume, characteristic of  $\text{NH}_3$  breakthrough, can be avoided by injecting ammonia below the stoichiometric requirement, i.e. operating the process in a starved ammonia mode.
4. Injection of  $\text{NH}_3$  before the fabric filter instead of the spray dryer resulted in a slight drop in utilization and earlier  $\text{NH}_3$  breakthrough at the outlet of the fabric filter. This could be due to the shorter residence time of the gas prior to the fabric filter.
5. The fabric filter pressure drop was not affected by the submicron  $\text{NH}_3/\text{SO}_2$  reaction product due to the buffering by the larger size  $\text{Ca}(\text{OH})_2/\text{SO}_2$  product in these short term tests. Longer term testing needs to be conducted.
6. Research on the regeneration of ammonia from the products is on-going to determine if the  $\text{NH}_3$  can be regenerated and under what conditions.

### ACKNOWLEDGMENT

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#### DISCLAIMER

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

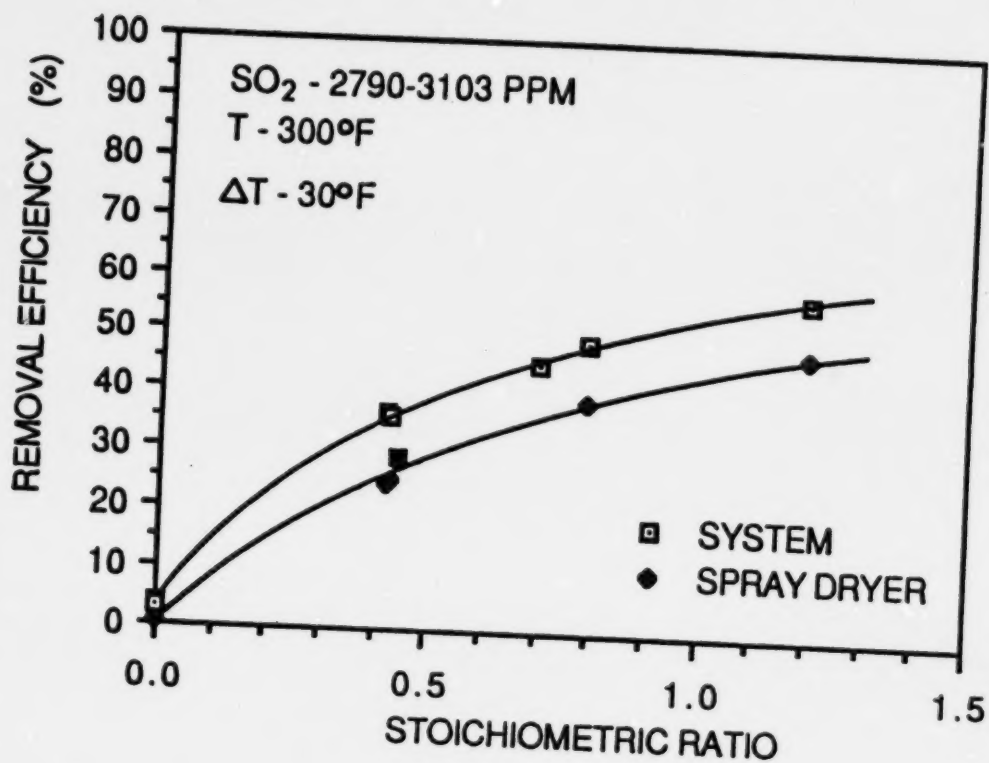


Figure 1. Performance of a  $\text{Ca}(\text{OH})_2$  Based Spray Dryer/Fabric Filter

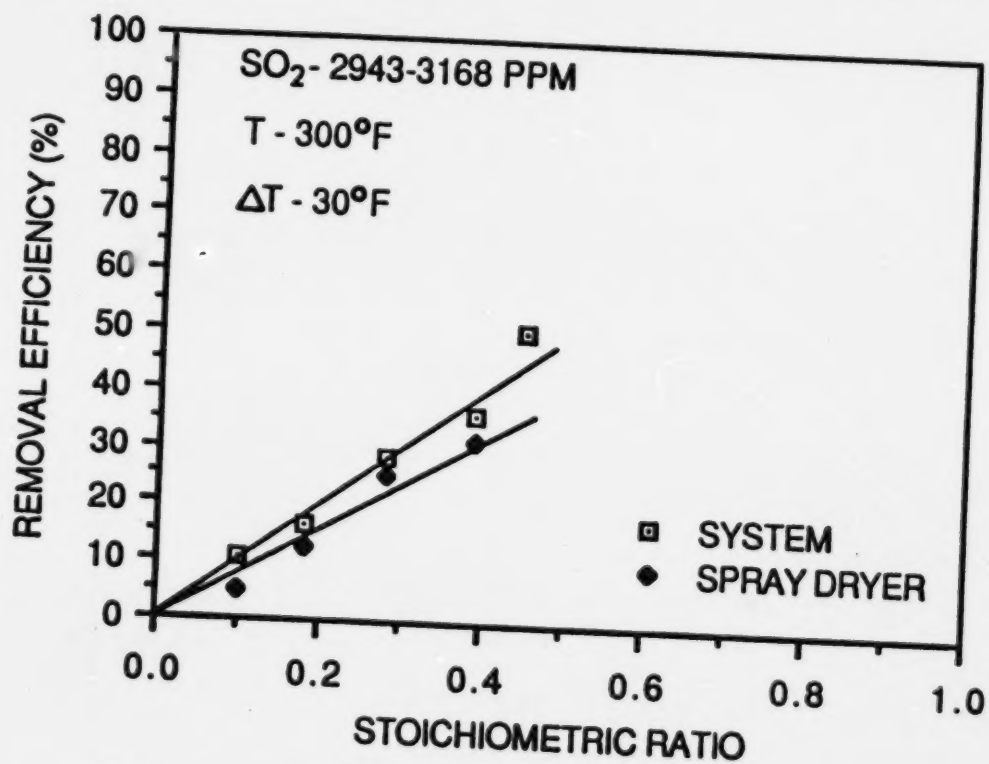


Figure 2. Performance of a Gaseous  $\text{NH}_3$  Injection System

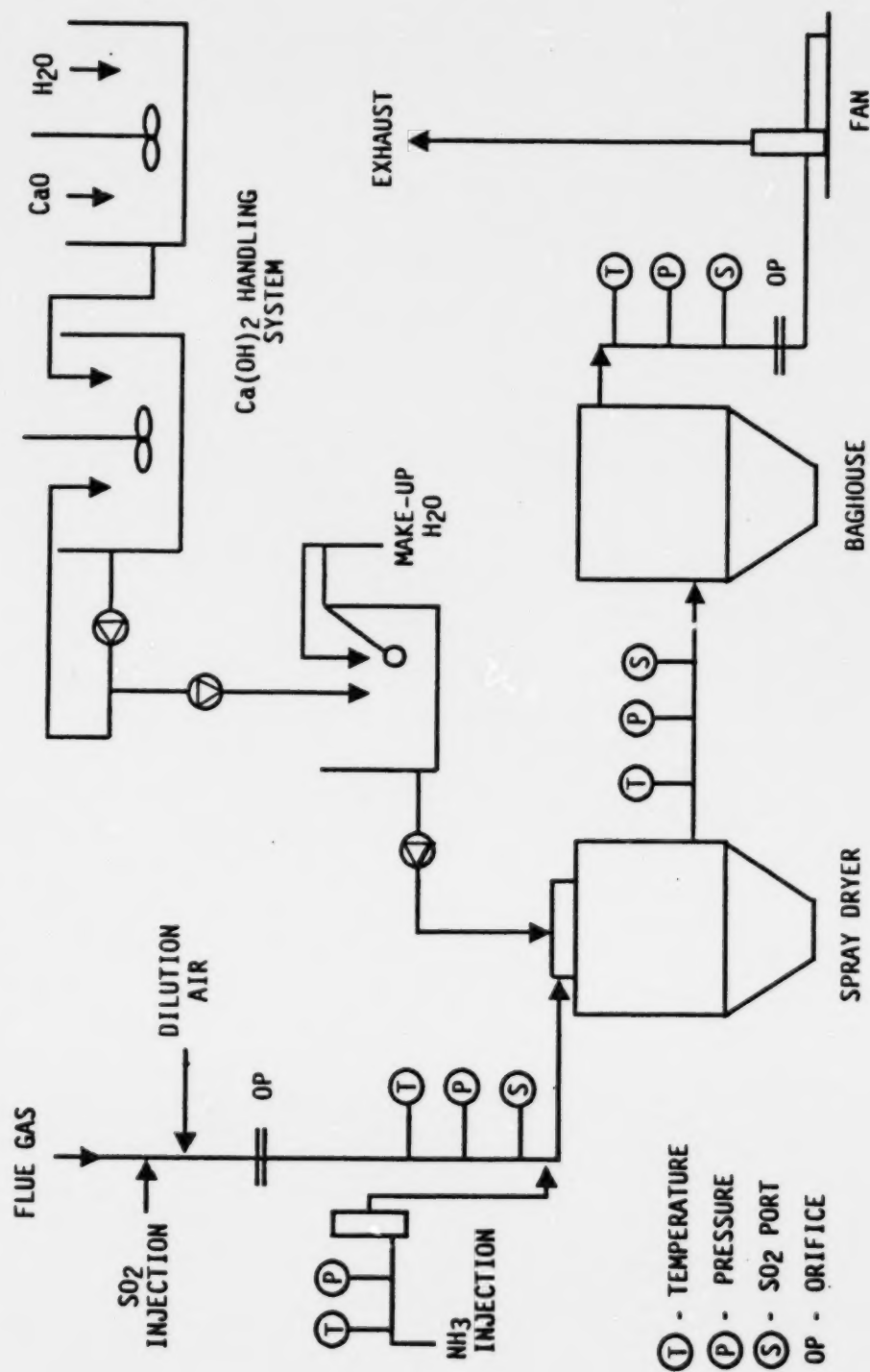


Figure 3. Schematic Diagram of the Spray Dryer/Fabric Filter Pilot Plant



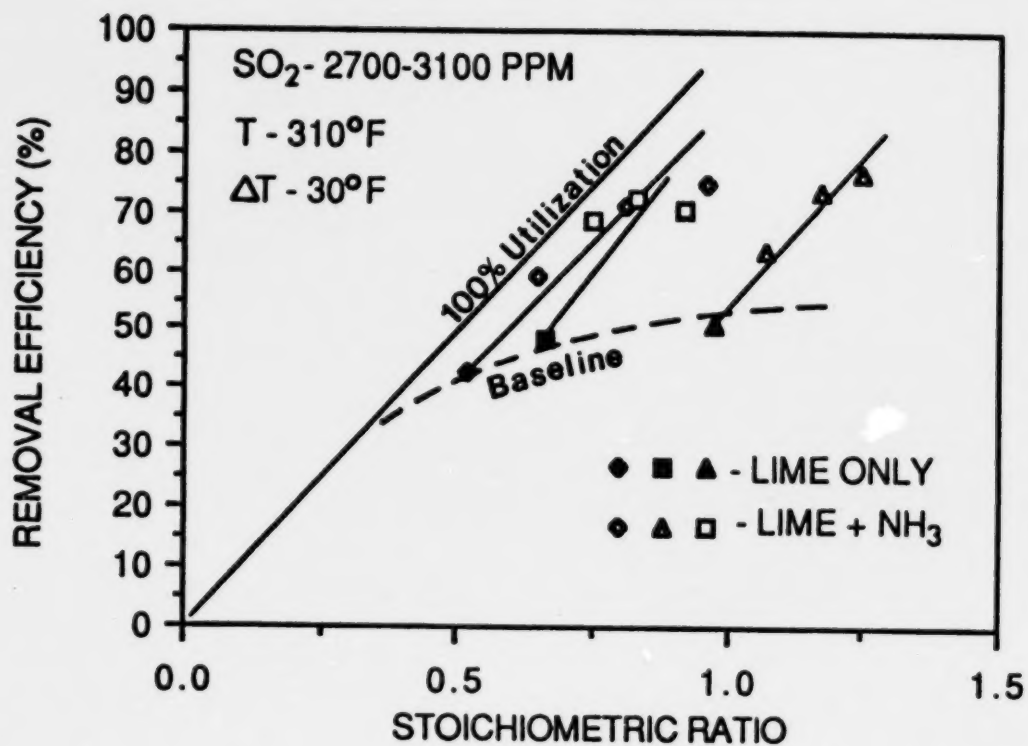


Figure 4. Performance of Spray Dryer

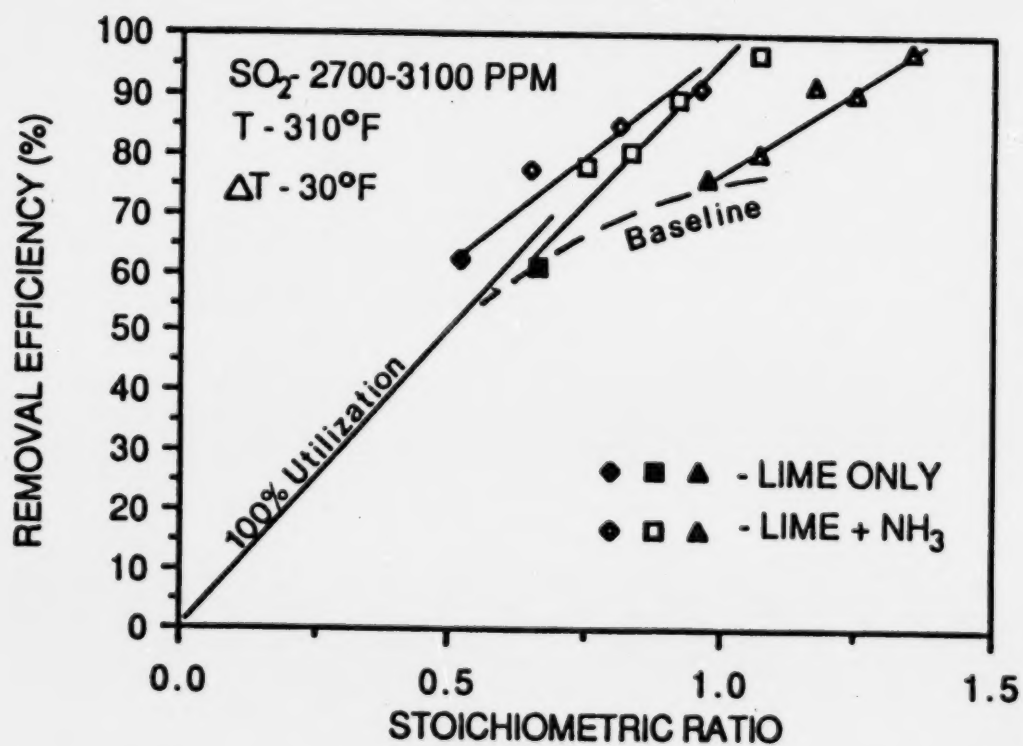


Figure 5. Overall Performance of Spray Dryer/Fabric Filter System

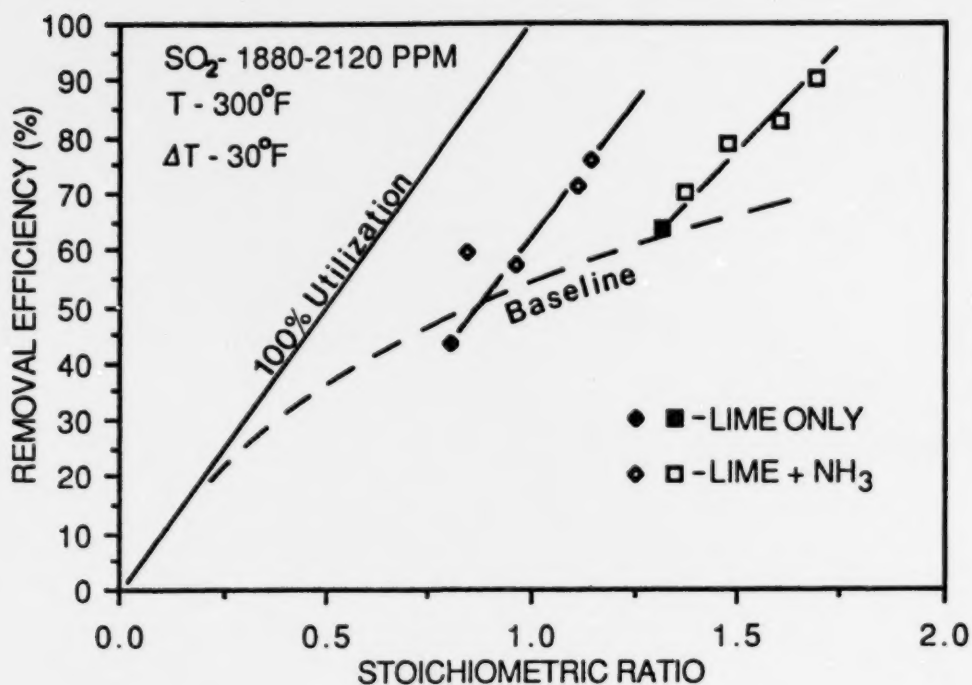


Figure 6. Performance of Spray Dryer

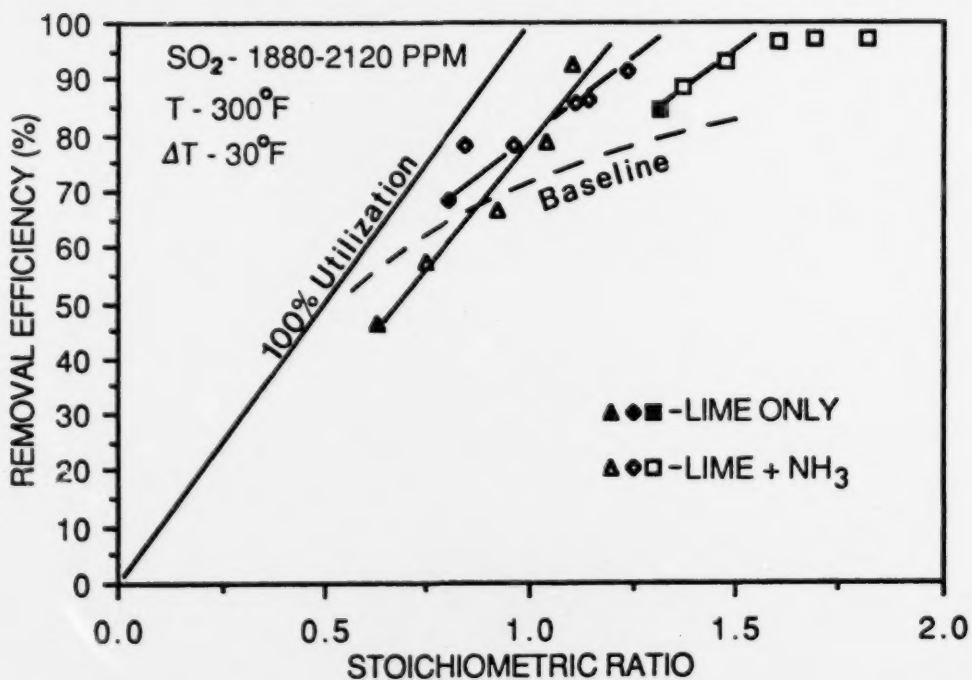


Figure 7. Overall Performance of Spray Dryer/Fabric Filter System

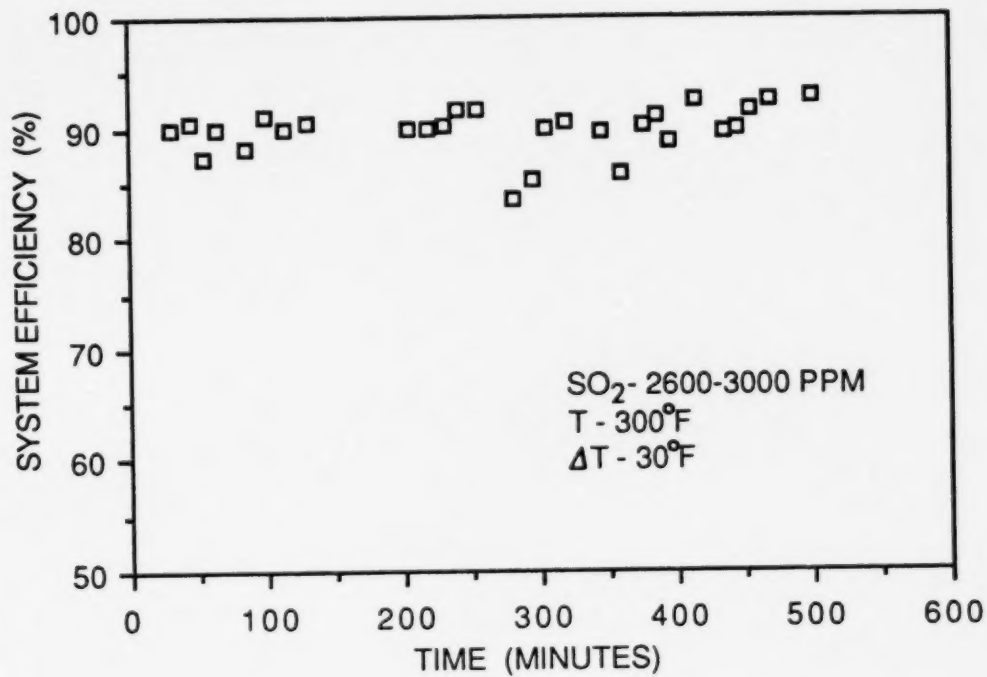


Figure 8. Overall Performance of System in Combined Injection Mode During Demonstration Tests

TABLE 1  
REACTION PRODUCT ANALYSES

|                                   |                    | UT<br>Flyash | Ca(OH) <sub>2</sub> | NH <sub>3</sub> | Combined<br>Waste |
|-----------------------------------|--------------------|--------------|---------------------|-----------------|-------------------|
| Water Content (%)                 | SD: --<br>BH: 0.8  |              | 5.2<br>0.7          | 7.3<br>0.0      | 3.7<br>3.0        |
| True Density (g/cm <sup>3</sup> ) | SD: --<br>BH: 2.04 |              | 2.67<br>1.68        | 1.96<br>1.82    | 2.56<br>2.93      |
| Bulk Density (g/cm <sup>3</sup> ) | SD: --<br>BH: 0.53 |              | 0.47<br>0.39        | 0.51<br>0.38    | 0.54<br>0.40      |
| Surface Area (m <sup>2</sup> /g)  | SD: --<br>BH: 7.88 |              | 9.19<br>8.50        | 7.82<br>4.07    | 8.00<br>6.64      |
| Ca (%)                            | SD: --<br>BH: neg. |              | 27.6<br>28.4        | 14.74<br>10.20  | 21.9<br>23.7      |
| NH <sub>3</sub> (% as N)          | SD: --<br>BH: 0.1  |              | ND<br>.05           | 3.9<br>12.3     | 9.5<br>5.9        |
| S (% at 820°F)                    | SD: --<br>BH: 1.2  |              | <.1<br>0.2          | <.1<br>2.3      | <.1<br>0.42       |
| S (% at 2600°F)                   | SD: --<br>BH: 1.5  |              | 7.4<br>8.6          | 10.9<br>11.7    | 10.3<br>10.4      |
| AMD (μm)                          | 1.0-2.0            | 55           | 0.2                 | --              |                   |
| σg                                | >10                | 6.3          | 8.6                 | --              |                   |

AMD = Aerodynamic Mean Diameter measured at Spray Dryer  
outlet by Cascade Impactor

σg = Geometric Standard Deviation of Particle Distribution



TABLE 2  
CONCENTRATION OF METALS IN THE TCLP EXTRACT

|          | RCRA<br>LIMIT<br>mg/l | % Allowable RCRA Limit      |                         |                         |                             |                                     |          |
|----------|-----------------------|-----------------------------|-------------------------|-------------------------|-----------------------------|-------------------------------------|----------|
|          |                       | Ca(OH) <sub>2</sub><br>ONLY | NH <sub>3</sub><br>ONLY | SPRAY DRYER<br>COMBINED | Ca(OH) <sub>2</sub><br>ONLY | BAGHOUSE<br>NH <sub>3</sub><br>ONLY | COMBINED |
| Arsenic  | 5.0                   | --                          | 0.35                    | --                      | 0.35                        | --                                  | --       |
| Barium   | 100.0                 | 1.0                         | 6.0                     | 3.0                     | 12.5                        | 7.5                                 | 3.0      |
| Cadmium  | 1.0                   | --                          | --                      | --                      | --                          | 47.0                                | --       |
| Chromium | 5.0                   | 1.75                        | --                      | --                      | 1.0                         | 0.75                                | 0.83     |
| Lead     | 5.0                   | --                          | --                      | --                      | --                          | --                                  | --       |
| Mercury  | 0.2                   | 1.0                         | 0.3                     | 0.1                     | 0.2                         | 0.05                                | 0.1      |
| Selenium | 1.0                   | --                          | 50                      | 30                      | 30                          | 180                                 | 150      |
| Silver   | 1.0                   | 3.4                         | 4.4                     | 3.2                     | 7.0                         | 3.2                                 | 2.0      |

UPDATE OF JOY/NIRO U.S.  
UTILITY SPRAY DRYER FGD SYSTEMS

J. R. Donnelly  
M. T. Quach  
JOY TECHNOLOGIES INC.  
Western Precipitation Division  
Monrovia, California 91016-3633

Steven H. Wolf  
Northern States Power Company  
Plant Engineering and Construction  
Sherburne County Unit 3 Construction

Glenn A. Fosher  
Pacific Power & Light  
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Presented at:

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FGD and Dry SO<sub>2</sub> Control Symposium  
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ABSTRACT

Since the last FGD Symposium in November 1986, two additional JOY/Niro utility spray dryer absorption (SDA) FGD systems have completed start-up and undergone preliminary performance testing. This brings to eight the number of U.S. systems in operation treating flue gas from the generation of 2,900 MW of electricity. These two units:

- ° Wyodak Generating Station, Unit 1 - 330 MW;
- ° Sherburne County, Unit 3 - 860 MW;

represent the first U.S. retrofit application of an SDA system upstream of an existing electrostatic precipitator and the largest SDA application in the world. Both units have undergone performance tests and operate in compliance with widely different state emission requirements. During start-up, several problem areas were identified and corrected.

The remaining six systems continue to operate with high availabilities meeting SO<sub>2</sub> and particulate emission requirements.

This paper presents a process description of the JOY/Niro SDA system, design parameters, and system descriptions of these two units. Performance test results are presented for these two units, start-up problems are reviewed, and solutions discussed. Availability data from all of the JOY/Niro U.S. utility SDA applications are presented.

## INTRODUCTION

Spray dryer absorption (SDA) was developed, during the mid- to late- 1970's, as an alternative to traditional wet lime and limestone scrubbers for utility flue gas desulfurization (FGD). It was perceived at the time that the simplicity of the SDA process would lead to higher availabilities and lower costs for primarily low sulfur coal applications. The SDA process was first employed commercially on coal-fired utility boilers in 1980. Since then, more than about 20,000 MW of generating capacity (equivalent) world-wide has been committed to the SDA technology with about 8,000 MW in operation. In addition, the SDA technology is being applied successfully to many smaller industrial and cogeneration plants, as well as waste incinerators for total acid gas, particulate, and trace element control. These operating plants have confirmed the basic design concepts, high availability, and low costs for the technology.

JOY TECHNOLOGIES INC. and A/S Niro Atomizer (JOY/Niro) are leaders in the development and application of SDA systems for all types of applications. Since 1977, when JOY and Niro first agreed to jointly develop and market the SDA technology in the United States, they have advanced the technology from the pilot plant stage, through full-scale utility and industrial demonstration plants, to full-scale applications achieving high SO<sub>2</sub> removal efficiencies (up to 98%) for low and high sulfur coals (up to 3.5%). JOY/Niro has received 10 orders for utility SDA systems in the United States representing approximately 4,000 MW. Eight of these systems have started-up and completed compliance and performance tests. Table I presents a list of these operating utility systems.

TABLE I. U.S. OPERATING INSTALLATIONS - UTILITY BOILERS.

| Installations<br>Name - Utility - Location                                                      | Plant Size<br>MWe | Gas Flow<br>acfm     | Type Coal<br>And Design<br>SO <sub>2</sub> Removal,% | Start-Up |
|-------------------------------------------------------------------------------------------------|-------------------|----------------------|------------------------------------------------------|----------|
| Riverside Generating Sta.Units 6 & 7<br>Northern States Power Company<br>Minneapolis, Minnesota | 110               | 645,000<br>@ 360°F   | Various<br>70-90                                     | Dec. '80 |
| Antelope Valley Station Unit 1<br>Basin Electric Power Cooperative<br>Beulah, North Dakota      | 450               | 2,055,000<br>@ 310°F | No. Dakota<br>Lignite<br>78                          | May '83  |
| Holcomb Station Unit 1<br>Sunflower Electric Cooperative<br>Holcomb, Kansas                     | 319               | 1,340,000<br>@ 253°F | Subbituminous<br>Powder River<br>Basin<br>80         | May '83  |
| Rawhide Energy Station Unit 1<br>Platte River Power Authority<br>Wellington, Colorado           | 250               | 1,356,000<br>@ 276°F | Subbituminous<br>Powder River<br>Basin<br>80         | Dec. '83 |



TABLE I. U.S. OPERATING INSTALLATIONS - UTILITY BOILERS.  
(Continued)

| Installations<br>Name - Utility - Location                                                 | Plant Size<br>MWe | Gas Flow<br>acfm     | Type Coal<br>And Design<br>SO <sub>2</sub> Removal, % | Start-Up  |
|--------------------------------------------------------------------------------------------|-------------------|----------------------|-------------------------------------------------------|-----------|
| Springerville Station Unit 1<br>Alamito Company<br>Springerville, Arizona                  | 350               | 1,660,000<br>@ 256°F | Subbituminous<br>New Mexico<br>82                     | June '85  |
| Antelope Valley Station Unit 2<br>Basin Electric Power Cooperative<br>Beulah, North Dakota | 450               | 2,055,000<br>@ 310°F | No. Dakota<br>Lignite<br>81                           | Oct. '85  |
| Wyodak Station Unit 1<br>Pacific Power & Light<br>Gillette, Wyoming                        | 330               | 2,260,000<br>@ 360°F | Subbituminous<br>Powder River<br>Basin<br>75          | Sept. '86 |
| Sherburne County Station Unit 3<br>Northern States Power Company<br>Becker, Minnesota      | 860               | 3,781,000<br>@ 308°F | Subbituminous<br>Montana Coal<br>75                   | Jan. '88  |

The first system came on-line at the Riverside Generating Station in December 1980, treating the combined flue gas streams from Units 6 and 7. The system was installed as a full-scale demonstration plant to prove out the technology and confirm scale-up factors from pilot scale. The system was extensively tested for more than three years on a variety of different coal combinations ranging in sulfur content from 0.5 percent up to 3.5 percent. The testing at Riverside demonstrated the ability of the SDA process to achieve high SO<sub>2</sub> removals, low emission rates, and high reliability. It also served as a test plant for selecting process design conditions and equipment for Northern States Power's Sherburne County Station, Unit 3. During most of the test program phase, and all of the commercial operating period, the single-chamber SDA system has operated at above 90 percent availability. The SDA system is currently in a stand-by mode with flue gas passing through the chamber, but no reagent feed, because the plant is firing a compliance coal.

From 1983 through 1985, five full-scale systems started-up, each equipped with large, multiple spray drying chambers. All of these units were grass roots plants and were equipped with reverse-air fabric filters for dust collection. Start-up problems and performance test results for these units have been reported at the two previous EPA/EPRI Symposiums on Flue Gas Desulfurization.

During the past two years, two additional units have started-up and undergone performance testing. These units, Wyodak Unit 1 and Sherburne County Unit 3 (SHERCO 3), present the first U.S. retrofit application and the world's largest SDA application respectively. Wyodak has had three spray dryer absorbers installed in front of existing electrostatic precipitators. SHERCO 3 is equipped with eight spray dryer absorbers, followed by three reverse-air fabric filters.

This paper presents a description of the JOY/Niro SDA process and design parameters for these two last units. Results of performance tests are presented and start-up problems are discussed. Availability data for all of the operating U.S. installations are presented.

## JOY/NIRO SYSTEM DESCRIPTION

The JOY/Niro SDA process combines Niro's experience in spray drying technology with JOY's experience in dust collection technology to yield an effective FGD process which results in a dry product for disposal. The SDA system consists of one or more spray dryers, one of more electrostatic precipitators or fabric filters for dust collection, and a reagent preparation system. Through development at pilot and demonstration plant scale as well as feed-back from early full-scale installations, several features have been incorporated in the standard system design. These include:

- Large spray dryers with compound gas dispersers;
- Single F-800 atomizer per chamber;
- Re-ash slurry recycle system;
- High feed slurry solids content;
- Two-point product discharge;
- Close approach to adiabatic saturation temperature operation;
- Nominal 10-12 second SDA residence time.

For the majority of U.S. utility applications where one to three hour SO<sub>2</sub> emissions-averaging times are mandated, feed preparation systems utilizing fresh lime slurry and a re-ash slurry concept are employed. Figure 1 is a simplified flow diagram showing this concept.

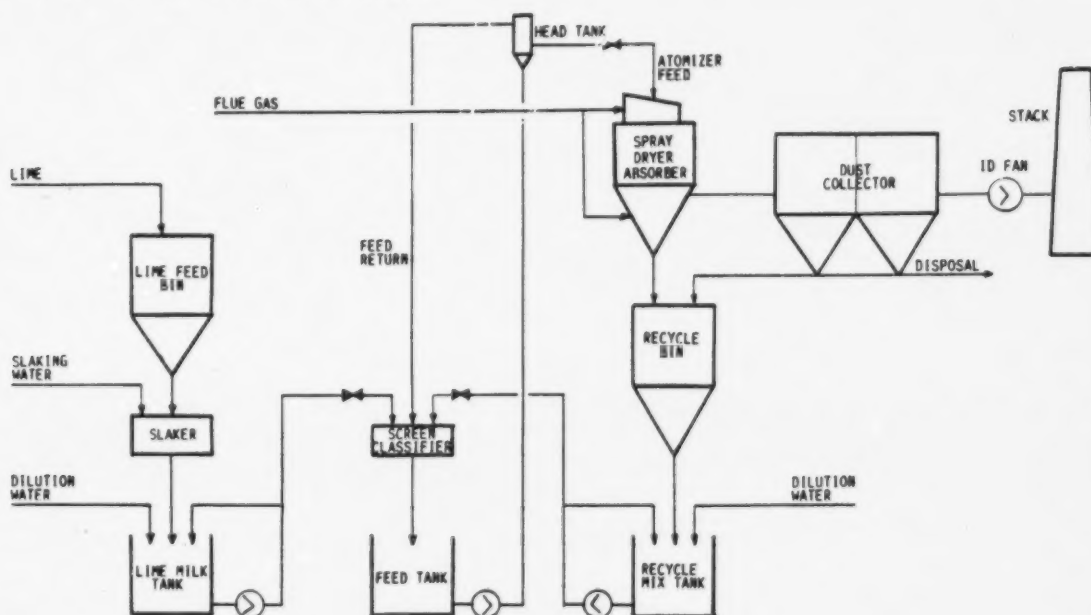


Figure 1. Simplified Flow Diagram.

Lime is fed from the lime bin to the slaker via a weigh belt. In the slaker the quicklime is converted to an hydrated lime slurry containing approximately 25 percent solids. When pebble lime is the reagent a ball mill slaker, which can grind up any grit present, is used. The slaked lime is then transferred to a sump or lime holding tank where it may be diluted using plant service or cooling tower blowdown water.

The dry product collected from the SDA or dust collector is transferred to a recycle bin for use in preparing a recycle slurry (re-ash). The product is fed from the recycle bin via a rotary or other type valve into a recycle mix tank

where a re-ash slurry containing 45-55 percent solids is prepared. Cooling tower blowdown or other low quality water is used to prepare the re-ash slurry.

The lime and re-ash slurries are then pumped through a screen to an agitated feed tank. The ratio between the lime and re-ash slurries is controlled to either a set outlet  $\text{SO}_2$  concentration or a percent  $\text{SO}_2$  removal. From the feed tank, the slurry is pumped to a constant level head tank where approximately 50 percent of the feed overflows and returns by gravity to the feed tank through the screen. The flow to the atomizer is controlled to maintain a constant SDA outlet temperature.

The high speed rotary atomizer converts the feed slurry into a multitude of discrete fine droplets (approximately 30 micron mean diameter) producing very high surface area for heat and mass transfer. The rotary atomizers used in these systems are designated F-800 atomizers and can accept up to 1200 horsepower drive motors.

The vertically flanged motor rotating at 3600 RPM drives a planetary gearbox via a tooth coupling. This planetary gearbox consists of a gear anulus, sun wheel and a number of planet wheels, each turning on a flexible spindle. This distributes tooth pressure uniformly on each planet gear and along the length of each gear mesh. Spindle speed can be altered by changing the tooth anulus, sun wheel and planet wheels. The atomizer can rotate in either direction, however, normal rotation is counter-clockwise.

The gearbox drives a flexible stainless steel shaft which operates between the first and second critical speeds. A self-lubricating guide bearing is mounted toward the lower end of the shaft to prevent the spindle from coming in contact with any stationary components as the spindle passes through the first critical speed.

The atomizer is equipped with independent dual-lubricating systems; one for the gearbox and one for the spindle bearings. Both systems include oil pumps, parallel oil filters, and monitors to ensure proper operation. The atomizer is designed for long life and low maintenance. For FGD applications, the atomizer is outfitted with a patented KN abrasion resistant wheel developed initially for mineral concentrate drying. Figure 2 shows an F-800 atomizer and the KN wheel.

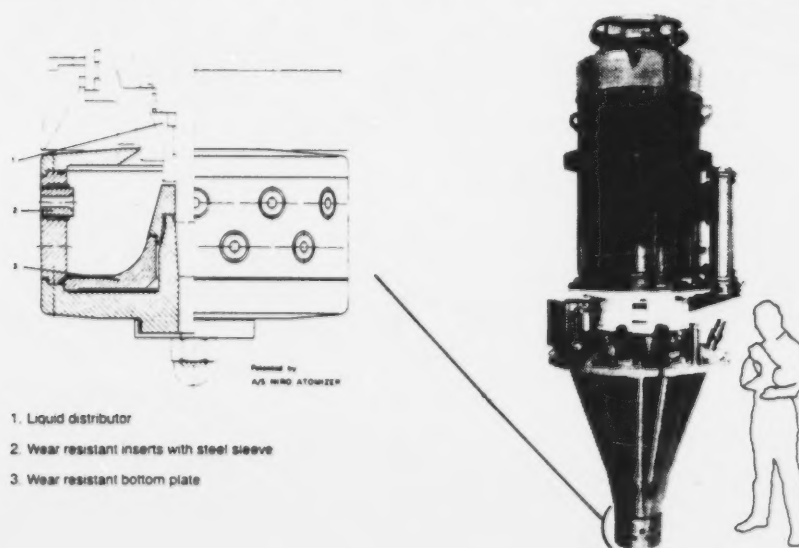


Figure 2. Rotary Atomizer and Patented KN Wheel.

The wheel consists of a wheel body made of stainless steel or more corrosion resistant alloy, a bottom wearplate, and inserts protruding into the wheel body to form a layer of slurry for protection of the inside of the wheel body when the wheel is rotating. The bottom plate and the inserts are made of abrasion-resistant material, e.g., silicon carbide, and are replaceable. The inserts can be rotated three or four times to extend their usable life.

Flue gas from the boiler is ducted to the flue gas cleaning system where it is split between the operating SDA's. In the case of Wyodak and SHERCO 3, a portion (4 to 30 percent) of the incoming from either before or after the air preheater is bypassed around the SDA's to provide reheat prior to the dust collectors. The flue gas entering each SDA is split such that approximately 60 percent enters through the roof gas disperser and the remainder through the central gas disperser. Vanes in the two gas dispersers impart a swirl to the incoming gas and assist in controlling the shape of the droplet cloud. Figure 3 illustrates a spray dryer absorber with a compound gas disperser, the position of the single rotary atomizer, and the two-point discharge design.

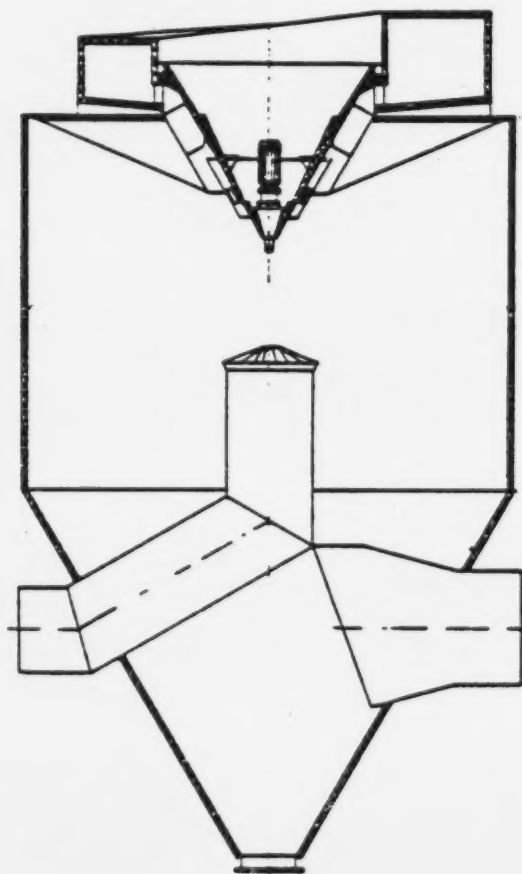


Figure 3. Spray Dryer with Compound Disperser.

The upper gas disperser includes a scroll type entry to maintain constant flue gas velocities all around the disperser and minimize fly ash dropout. The roof gas disperser includes two sets of adjustable vanes whereas the central gas disperser has one. Figure 4 shows these vanes.



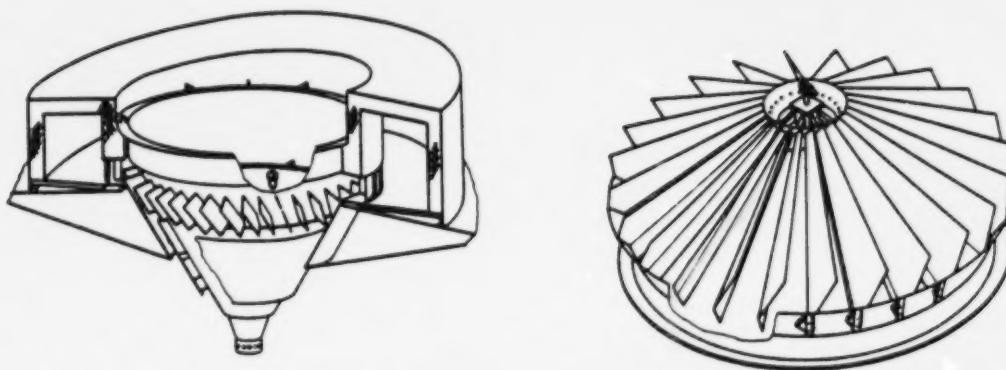


Figure 4. Roof and Central Gas Disperser Vanes.

Initial adjustment of the gas flow split and disperser vanes ensures efficient mixing of the flue gas and absorbent in a narrow zone around the atomizer. This, in turn, results in good contact between the incoming acid gases ( $\text{SO}_2$ ,  $\text{SO}_3$ ) and alkaline feed, yielding high acid gas removal and adequate drying of the feed slurry. The flue gas exits the dryer via a side discharge, carrying the major portion of the dried absorbent and reaction products to the dust collector. During normal operation, about five percent of the dried material drops out to the SDA bottom. During upset conditions, the two-point discharge ensures an open gas flow path even if wall deposits occur and fall to the bottom of the chamber. The discharge from the chamber is equipped with a lump breaker to ensure that collected material can be conveyed away.

In the dust collector, fly ash, absorbent and reaction products are removed from the flue gas and transported either to the recycle bin or to disposal. Additional  $\text{SO}_2$  removal takes place in the dust collector. The split in sulfur dioxide removal between the spray dryer and the dust collector varies with operating conditions and type of dust collector used. When a bag filter is used, typically between 60 and 70 percent of the absorption takes place in the spray dryer. For an ESP, this value increases to 80 to 90 percent of the total.

#### WYODAK SYSTEM

The Wyodak station is the first JOY/Niro utility retrofit application and the first full-scale commercial system to use an electrostatic precipitator as a dust collector. Prior to award of the contract for this system, Pacific Power & Light, JOY, and Niro conducted an extensive pilot plant program at Niro's research facilities in Copenhagen, Denmark. The purpose of the test program was to characterize the performance of the SDA system and particularly the three-field pilot electrostatic precipitators under Wyodak conditions.

After award of the contract in January 1984, a second pilot plant program was conducted at Niro's facilities. The purposes of this second test program were to verify process conditions for the retrofit application and to determine if any detrimental effects of long term operation would be observed on the performance of the precipitator. The test program included operating with and without the SDA in service and under transition conditions. The results of the test program indicated that the required  $\text{SO}_2$  and particulate removal efficiencies could be achieved under all operating conditions. The expected ESP outlet grain loading was lower with the SDA's in service than without. Table II presents design data for the Wyodak SDA system.

TABLE II. WYODAK DESIGN PARAMETERS.

|                                                |                        |
|------------------------------------------------|------------------------|
| Gas Flow                                       | 2,260,000 acfm @ 360°F |
| <u>Spray Dryers</u>                            |                        |
| number                                         | 3                      |
| size, ft 0 x ft                                | 49 x 39                |
| gas residence time, sec                        | 10                     |
| exit temperature, °F                           | 151                    |
| approach to Ad Sat, °F                         | 20                     |
| feed solids, %                                 | 40                     |
| atomizer motor, Hp                             | 1100                   |
| <u>Dust Collector</u>                          |                        |
| inlet temperature, °F                          | 160                    |
| type                                           | ESP                    |
| electrode system                               | Rigid Frame            |
| fields                                         | 5 + 1 Vacant           |
| approx SCA, ft <sup>2</sup> /1000 acfm         | 650                    |
| <u>Slakers</u>                                 |                        |
| lime type                                      | Pebble Quicklime       |
| slaker type                                    | Ball Mill              |
| capacity, TPH                                  | 2 x 7                  |
| Reheat                                         | Warm Bypass            |
| SO <sub>2</sub> inlet design, ppm              | 730                    |
| removal %, lbs/10 <sup>6</sup> BTU             | up to 75               |
| emissions, lbs/10 <sup>6</sup> BTU             | 0.5                    |
| Particulate emissions, lbs/10 <sup>6</sup> BTU | 0.1                    |

The system is designed for normally operating with three spray dryers on-line with approximately five percent flue gas bypass to reheat the gas before the ESP's. The system can also operate at design performance conditions with two spray dryers on-line when the inlet SO<sub>2</sub> concentration is below 700 ppm while bypassing approximately 20 percent of the flue gas. The design SO<sub>2</sub> removal varies with inlet SO<sub>2</sub> concentration to maintain an emission level of 0.5 lbs/10<sup>6</sup>BTU.

The SDA system started-up in September 1986, and quickly achieved full load operation while maintaining the required SO<sub>2</sub> emissions and stack opacity. However, during the first winter of operation, problems were encountered with condensation, corrosion in the ESP inlet and outlet ducts, and the ESP ash conveying system suffered severe plugging due to water condensing in the ESP. Early in the engineering phase of the project, a decision was made to add wall insulation to the outside of the ESP, and not insulate the ESP inlet and outlet ducts. As it turned out, this decision was in error. In 1987, JOY added additional insulation to the ESP inlet and outlet ducts, nozzles, and to the area between the ESP chambers and the breaching between the stack and stack liners. This has apparently resolved condensation problems in the ESPs, as evidenced by satisfactory operation since the spring of 1988.

The insulation also improved operation of the ash conveying system, but problems were still encountered with achieving conveying capacity. By adding some additional air venting to the conveying system and reducing the speed of the conveying air blowers, the capacity has been increased to above the design capacity. This increase in conveying capacity yields a more reliable recycle system which results in better control of recycle slurry solids.

In November 1986, preliminary performance tests and state compliance tests were carried out with three SDA's in service at full load. Inlet flue gas temperatures were approximately 45°F below design and the adiabatic temperature was about 5°F above design, resulting in lower spray down across the SDA's. The inlet SO<sub>2</sub> concentration averaged about 525 ppm and the outlet 195 ppm corresponding to a removal efficiency of 63 percent and an emissions rate of 0.47 lbs/10<sup>6</sup>BTU. The lime consumption was 11 percent lower than the guaranteed value. Before and after the preliminary performance tests, episodes would occur where deposits were formed in the SDA chambers. The cause of these episodes was difficult to determine and required a detailed investigation by Niro. It was found that a combination of a mechanical and a chemical problems were causing the deposits.

The mechanical problem was due to improper setting of the gas flow split between the roof and central gas dispersers during initial start-up. The split was almost even, with about 50 percent of the flue gas going to the central gas disperser and 50 percent to the roof gas disperser. This resulted in lower SDA pressure drops and allowed higher than design gas flows through the chambers. At the higher gas flows and subsequently lower residence time and improper gas dispersion, drying was inadequate resulting in wall deposits. The split has been reset to the proper values and operation has been substantially improved.

The second problem would occur at low inlet SO<sub>2</sub> concentrations and high unit loads where little lime was added to the feed slurry and where recycle rates would become very high. Dissolved solids introduced into the system from the fly ash and dilution water would concentrate at very high levels under these conditions. This would result in a feed slurry containing significant amounts of dissolved solids and a reduced amount of suspended solids for a given total feed solids content. Drying of the feed slurry would be impaired, resulting in wet material hitting the walls and building up. The solutions to this problem have been to modify the control logic to maintain a minimum concentration of lime in the feed slurry and to raise the spray dryer outlet temperature when conditions of low feed suspended solids and lime content occur.

These solutions have resolved the problems which caused unreliable system operation and led to the initiation of a 2000 hour reliability run in June 1988. The reliability run was completed in August with a system reliability of greater than 98 percent during this period. After completion of the reliability run, a series of performance tests were conducted at full, 50 percent and 75 percent boiler loads. Table III presents preliminary results from tests at full load. Seven test runs were carried out at full load over a three day period in early September 1988.

TABLE III. WYODAK PERFORMANCE TEST RESULTS - PRELIMINARY.

|                                                      | Measured<br>Average | Design or<br>Guarantee Value |
|------------------------------------------------------|---------------------|------------------------------|
| Inlet Gas Flow, acfm                                 | 894,560             | 1,209,000                    |
| Inlet Temperature, °F                                | 299                 | 360                          |
| Inlet Particulate, gr/acf                            | 1.72                | 2.0                          |
| Inlet SO <sub>2</sub> , ppmvd                        | 587                 | 700                          |
| Outlet SO <sub>2</sub> , ppmvd                       | 197                 | 219                          |
| Outlet SO <sub>2</sub> , lbs/10 <sup>6</sup> BTU     | 0.45                | 0.5                          |
| SO <sub>2</sub> Removal, %                           | 66.4                | 62.7                         |
| Dust Emissions, lbs/10 <sup>6</sup> BTU<br>, gr/dscf | <0.01<br>0.0045     | 0.1                          |
| System Pressure drop, in. W.G                        | 8.1                 | 10.5                         |
| Stoichiometric Ratio                                 | 0.43                | 0.60                         |



The plant was operated at close to full load during the test period. Inlet  $\text{SO}_2$  concentrations averaged 587 ppmvd and ranged from 480 to 710 ppmvd. The actual  $\text{SO}_2$  removal was higher than required and resulted in  $\text{SO}_2$  emissions averaging 0.45 lbs/10<sup>6</sup>BTU at a stoichiometric ratio of 0.43 based on inlet  $\text{SO}_2$  to the system and the lime weighbelt feeder. This compares with a guarantee value of 0.60 at these inlet conditions. The dust emissions were well below the guarantee of 0.1 lbs/10<sup>6</sup>BTU and the values fit well with JOY's prediction for these operating conditions. System pressure drop corrected to full load conditions was below the guarantee value. The plant is now in period three of the performance test program, 4000 hours of on-load operation, prior to the final performance tests.

Two problems continue to occur at this installation. One is due to the wide variation in the sulfur content of the coal delivered to the plant. This coal produces flue gas  $\text{SO}_2$  concentrations which often exceed the design maximum inlet concentration (700 ppm) by more than 50 ppm, and in one case, has gone as high as 1400 ppm. At the higher than design  $\text{SO}_2$  concentrations, the reagent preparation system cannot produce enough feed slurry (specifically the lime fraction) under normal design operating procedures. To handle these high  $\text{SO}_2$  levels, both slakers are operated with one dumping slaked lime into a floor sump where it is then transferred to the feed tank by a sump pump. Efforts are being made to lower the peak sulfur content of the plant coal.

An additional problem that has not been fully resolved is a high wear rate on the rotary atomizer wheel inserts. Due to the higher than expected abrasive nature of the feed slurry, the inserts are only achieving approximately 200 hours per wear track. Niro has recently supplied a harder material insert which is expected to give much lower wear rates. The initial set of new inserts have given longer life but have been shown to have lower flow capacities resulting in other problems. Niro is currently redesigning these harder material inserts to increase their flow capacities.

The start-up and de-bugging of the Wyodak SDA system has taken longer than planned. New insights have been gained on the effects of feed slurry chemical and physical properties on the drying and absorption phenomenon taking place in the spray dryer. This knowledge will assist in improved process design for future applications. The Wyodak system is operating satisfactorily and should meet or exceed the design availability of 90 percent.

### SHERBURNE COUNTY UNIT 3

The 860 MW (gross) Unit 3 at Northern State Power's Sherburne County (SHERCO) station started-up in the summer of 1987, and went commercial on November 1, 1987. The unit is equipped with the world's largest SDA-FGD system and incorporates equipment and design features developed through testing at the JOY/Niro installation at NSP's Riverside Generating Station. Three separate one month test programs were conducted while SHERCO's design coal was fired in Riverside boilers. NSP conducted equipment tests and stationed engineering personnel at Riverside to monitor equipment and system performance. This effort resulted in one of the smoothest start-ups of any SDA system.

The SHERCO 3 SDA system includes eight spray dryers and three sixteen compartment reverse air fabric filters. Figure 5 shows a plan view of the SDA system.



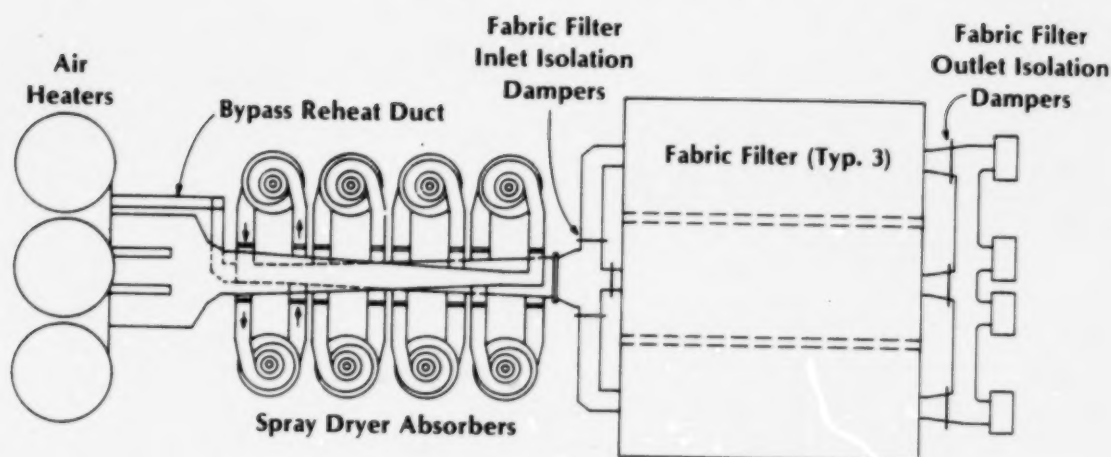


Figure 5. Plan View.

Any six of the eight spray dryers and 42 of the 48 fabric filter compartments can handle MCR gas flow volumes. Reheat between the spray dryers and fabric filters is by hot flue gas taken from before the air preheaters. Table IV presents design data for the SHERCO 3 SDA systems.

TABLE IV. SHERCO 3 DESIGN PARAMETERS.

|                                                |                           |
|------------------------------------------------|---------------------------|
| Gas Flow                                       | 3,718,000 acfm @ 308°F    |
| <b>Spray Dryers</b>                            |                           |
| number                                         | 6 + 2                     |
| size, ft 0 x ft                                | 46 x 36                   |
| gas residence time, sec                        | 9                         |
| exit temperature, °F                           | 143                       |
| approach to Ad Sat, °F                         | 18                        |
| feed solids, %                                 | 48                        |
| atomizer motor, Hp                             | 900                       |
| <b>Dust Collector</b>                          |                           |
| inlet temperature, °F                          | 165                       |
| type                                           | Reverse Air Fabric Filter |
| number                                         | 3                         |
| compartments/filter                            | 16                        |
| bags/compartments                              | 384                       |
| air-to-cloth ratio                             |                           |
| gross/net net, ft/min                          | 2.0/2.3                   |
| <b>Slakers</b>                                 |                           |
| lime type                                      | Pebble                    |
| slaker type                                    | Ball Mill                 |
| capacity, TPH                                  | 2 x 15                    |
| Reheat                                         | Hot Bypass                |
| SO <sub>2</sub> inlet design, ppmvd            | 750 (range 360-2070)      |
| removal percent                                | 70 (range 70-90)          |
| emissions, lbs/10 <sup>6</sup> BTU             | 0.6                       |
| Particulate emissions, lbs/10 <sup>6</sup> BTU | 0.03                      |

The fabric filters at SHERCO 3 are specifically designed for low temperature operation downstream from a spray dryer. Specific design features for this application fall into the following categories:

- ° fabric filter insulation;
- ° ambient air inleakage;
- ° spot coating application.

The insulation for the SHERCO 3 fabric filters is five inches thick on all exterior surfaces and three inches thick between compartments and between each compartment and the gas plenum. Fabric filter wall stiffeners are located internally to allow the insulation to be placed directly against the outside wall. To minimize heat loss from the hopper crotch area, insulation is installed on crotch areas and on the supporting columns.

Ambient air inleakage can cause localized corrosion. To minimize air inleakage around the fabric filter doors at SHERCO 3, the doors are separated into inspection and maintenance doors. The inspection doors are small, hinged doors fitted with quick-opening latches--one door per compartment. Larger bolted maintenance doors are also present and are designed to have tighter seals than the inspection doors.

As a final protection against corrosion, the internal surfaces of the fabric filter compartments were painted with a corrosion resistant coating, selected based on tests performed in the Riverside fabric filter. Two types of coatings were used; epoxy resins and vinyl ester resins. The epoxy material was used on interior walls and the roof while the vinyl material was used on walls to the exterior or "colder" surfaces.

Prior to start-up of the unit, NSP elected to pre-coat or "seed" a portion of the fabric filter. Ten compartments in one of the three fabric filters were designated for start-up of the unit and were seeded. This was accomplished by trucking in 200 tons of fly ash from an area plant which burns similar coal. The fly ash was blown into different elevations of the inlet duct of the fabric filter with the I.D. fans in service. These ten compartments were put into service within 30 minutes of the initial coal firing of the boiler, resulting in a chimney opacity of less than one percent within ten minutes. These compartments were used for the remainder of the unit start-up to support coal firing and checkout of the pulverizers. During oil firing, the fabric filters were bypassed.

After the boiler, pulverizers, and associated equipment had been sufficiently tested, the unit went into its first extensive operation. At this time, the flue gas temperatures were high enough (above 240°F) to support initial spray drying and sulfur removal on the unit. Initial spraying was done with lime slurry alone. Outlet temperatures on the SDA's were taken down as low as 180°F. The remainder of the fabric filter compartments were put into service filtering spray dried lime slurry and fly ash from the flue gas. With this combination, the chimney opacity was lowered to less than one percent after about 30 minutes of filtering through the virgin cloth.

During the next period of testing, recycle material was introduced and sprayed as often as possible to de-bug material handling aspects of the operation with recycle. The majority of the spray drying and sulfur removal was still accomplished with lime slurry only. This led to a major problem when one of two 2500 ton disposal silos set up with the high calcium product. NSP found that the silo had large areas or layers where a reaction had occurred, and the normally dry powder had become hard and "chalk like." Both silos were eventually cleared of this material, but only after an extensive effort. Emphasis was put on the use of recycle slurry in all future spray drying operations to minimize the use of lime slurry only and eliminate any extended storage of high calcium product.

Overall, the system operates very well and has not suffered any major problems which interfere with plant operation. The plant has completed compliance testing, preliminary performance tests, a 60-day reliability run and final performance testing. The reliability run was completed without any unit derates attributable to the FGD process or equipment problems. Table V presents the results of performance tests conducted at MCR conditions.

TABLE V. PERFORMANCE TEST RESULTS - SHERBURNE COUNTY UNIT 3.

| Time Period                                       | Initial Test Period<br>Jan. 26-27, 1988<br>Average | Final Test Period<br>August 9, 1988<br>Average | Design or<br>Guarantee<br>Value |
|---------------------------------------------------|----------------------------------------------------|------------------------------------------------|---------------------------------|
| Gross Generation, MW                              | 889                                                | 904                                            | 860                             |
| Flue Gas Flow, acfm                               | 3,360,000                                          | 3,450,000                                      |                                 |
| SDA Inlet Temp., °F                               | 273                                                | 285                                            |                                 |
| Inlet SO <sub>2</sub> , ppm <sub>dv</sub>         | 510                                                | 420                                            |                                 |
| SO <sub>2</sub> Removal, %                        | 72.4                                               | 74.9                                           | 72.4                            |
| Particulate Removal, %                            | 99.4                                               | 99.4                                           | ----                            |
| Particulate Emissions,<br>lbs/10 <sup>6</sup> Btu | 0.021                                              | 0.024                                          | 0.03                            |
| System Pressure Drop, "WG                         | 11.3                                               | 9.8                                            | 13.9                            |
| Stoichiometric Ratio                              | 0.43                                               | 0.49                                           | 0.67                            |

As is shown on the table, the SDA system has met performance guarantees for SO<sub>2</sub> removal, lime consumption, particulate emissions and system pressure drop. The performance tests also included 55 percent and 25 percent load tests. Performance guarantees were also met at these loads.

Problems encountered during the first 11 months of commercial operation of the SHERCO 3 FGD system were in the areas of atomizer vibration, FGD product handling and transport, SDA bottom lumpbreaker design, and the recycle slurry system solids feed and mixing design.

Atomizer vibrations were occurring after 100 to 300 hours of operation requiring change out of the running atomizer with a spare. The atomizer wheel was then pulled off and cleaned along with the liquid distributor to remove deposits or "ribs" which were causing the high vibration. A number of changes have been tested with a combination of modified liquid distributors and wheel components to lengthen the period of operation prior to vibration causing the need to remove the atomizer for cleaning. Presently, this time period is over 500 hours. The wheel nozzle wear rates at SHERCO 3 are very good. To date, the nozzles run over 600 hours before a rotation or change is required. Niro is continuing to investigate ways to lengthen the period between cleanings to match that required for nozzle maintenance.

The FGD product/solids handling system is a dilute phase pneumatic transport with pressure feeders under each of the fabric filter hoppers. A number of problems surfaced during initial operation of this system, especially during cold winter months. The most annoying problems were pluggage of the pressure feeder vent lines and partial pluggage or buildups in the feeders themselves. This pluggage has been attributed to the moisture laden flue gas entering the feeders or vent line along with the FGD product and condensing on cold surfaces. The FGD product would then stick and cause the pluggage. NSP has elected to install heat in the fabric filter enclosure which was not included in the original design. A number of other changes to the design were tested and implemented which include re-routing some of the transport and fluidizing air to the vent lines and different areas of the pressure feeders to keep them purged of any flue gas during



fill and vent cycles. Air supplied to the pressure feeder for fluidization and emptying of the feeder was also relocated to decrease the time required for all of the material to leave the pressure feeder and enter the conveying pipe. Some minimal changes were made to the automated control to shorten the time that the product sat stagnant in the pressure feeders. All changes have shown promising results and NSP is anxious to see how the system performs during the upcoming winter months.

Another problem identified during the first few months concerned design of the lumpbreakers installed at the bottom of the SDA chambers. A lumpbreaker was built and installed on the Riverside spray dryer during testing there. This design was modified and used at SHERCO 3. The lumpbreakers initially were attempting to break up too large of pieces of deposits which shed from the walls, causing the lumpbreakers to stall. This resulted in the chamber bottoms plugging and manual removal of the material was required. The lumpbreakers were modified by shortening the length of the rotating teeth, opening the clearances in the lumpbreaker and installing a sizing screen on the discharge. The redesign has operated successfully for over four months and all remaining lumpbreakers are in the process of being changed.

The last problem which required revision was equipment used to feed dry recycle material to the recycle slurry mix tank. The original design used a variable speed drag chain conveyor to meter material into recycle tank based on density measured by a sonic density meter. It was found that the drag chain could not control the material flow as the fluidized product would rush past the flights of the conveyor. Even after clearances in the conveyor were tightened up as much as possible flooding continued. The short term fix was to install a positioner on the silo outlet isolation knife gate valve to throttle the flow of recycle powder to the tank. At this time, it was also identified that the response and reliability of the density instrument was insufficient. It was found that throttling a valve based on the agitator power resulted in an acceptable slurry density control loop. An additional V-notch gate valve designed for throttling was installed and the slurry density control was changed to incorporate the use of agitator horsepower. This system now operates satisfactorily.

The chutework going to the recycle tank and the vent line for the moist, dusty air have had plugging problems. The agitator was lowered further into the tank to allow for a lower normal operating level. A portion of the chutework immediately above the tank has been lined with teflon to help solve the frequent pluggage at that location. Better access and inspection to the chutework has also been provided. These changes have been installed for almost two months and have addressed the problem of the wet/dry interface and chute pluggage at the top of the recycle slurry tank. The pluggage of the vent line with sticky material is still bothersome, causing the tank to become pressurized and dusty air escaping to the immediate surroundings. Work is continuing to resolve this problem.

#### SDA SYSTEM AVAILABILITY

In spite of the problems just discussed, JOY/Niro SDA systems continue to prove to be highly reliable FGD systems for utility boilers. The redundancy built into the systems and the high reliability of the spray dryer, atomizer and dust collectors, very seldom result in boiler downtime or curtailment of load. Table VI presents reported availability data for U.S. utility operating systems.



TABLE VI. SDA SYSTEM AVAILABILITY.

| Plant           | Antelope Valley<br>Units 1 & 2 | Holcomb<br>Unit 1 | Rawhide<br>Unit 1 | Springerville<br>Unit 1 | Wyodak<br>Unit 1 | Sherburne<br>Unit 3 |
|-----------------|--------------------------------|-------------------|-------------------|-------------------------|------------------|---------------------|
| Availability, % | 96+                            | Virtually<br>100  | 99+               | 97.7                    | 98+              | 99+                 |
| From            | Nov. 85                        | Jan. 85           | Jan. 86           | Jan. 86                 | Jan. 88          | Nov. 87             |

As is shown, all of the U.S. operating installations are achieving very high availabilities ranging from greater than 96 percent to 100 percent. These high availabilities extend from the newest installation to the oldest one.

The Antelope Valley Station units continue to show improved performance over time. This is due to system modifications and increased operator and maintenance personnel experience. Unit 1 has achieved greater than 99 percent availability over the past two years.

Holcomb Unit 1 continues to have the highest availability of all of the operating systems, 100 percent. In the five years since the unit was declared commercial, the boiler has not had to reduce load because of FGD system problems. Holcomb has a well developed set of operation and maintenance procedures and personnel who see that problems are solved before they become serious.

Rawhide Unit 1 has had an overall power plant availability factor of 91.8 percent with a capacity factor of 87.3 percent for the 12 month period ending September 30, 1988. For the most part, the SDA system has had no impact on the overall plant availability.

Springerville's availability is based on the percentage of the time they experience exceedences in emissions due to the SDA system. The value was 93.6 percent for the first six month reporting period and has increased every year since then to it's current level.

The availabilities for the Wyodak and SHERCO units are high, especially considering how new these applications are. The availability of the SHERCO 3 FGD equipment has been exceptional. Only one occurrence is recorded where availability of SDAs and associated equipment caused a unit derate. This represents an availability of 99.87 percent. This reflects not only a design which is free of major flaws but also a tremendous effort on the part of the operating staff who were working with a new technology and process.

## CONCLUSIONS

Currently, there are seven JOY/Niro U.S. utility SDA systems in operation. These systems are typically installed on new boilers and employ a fabric filter as the dust collector. Wyodak is the exception in that the SDA's were successfully retrofitted between the existing boiler and precipitators.

The last two units to come on-line (Wyodak and SHERCO 3) have both successfully completed compliance and performance tests. Today, they are operating satisfactorily at high availabilities and with diminishing problem areas.

All of the operating installations are operating at very high availabilities (up to 100 percent). Problem areas have, for the most part, been resolved through the efforts of the plant personnel and JOY/Niro.

The annual SDA users meeting has become a forum where there is an open and frank discussion of problems and solutions. The information transferred at these meetings has helped make this relatively new technology the highly reliable process it is today.

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UPDATE ON SPRAY DRYER FGD EXPERIENCE  
IN EUROPE AND IN THE PEOPLE'S REPUBLIC OF CHINA

by: Karsten Felsvang  
A/S Niro Atomizer  
Copenhagen, Denmark

Ragnar Hahn  
AB Aroskraft  
Västerås, Sweden

Dr. Peter Necker  
Neckarwerke Elektrizitätsversorgung AG  
Esslingen, W. Germany

M. Novak  
VKG Verbundkraft Elektrizitätswerke GmbH  
Vienna, Austria

Yang Qing  
China Aeronautical Project and Design Institute  
Beijing, China

N. Raffelsberger  
EVN Energie-Versorgung Niederösterreich  
Vienna, Austria

Erik Schumacher  
I/S Midtkraft  
Aarhus, Denmark

Hans Spannbauer  
Salzburger Stadtwerke  
Salzburg, Austria

ABSTRACT

The use of spray dryer absorbers for FGD was pioneered in the United States in the late 70's. Although the use of spray dryer absorbers in Europe was introduced later, there is now gained more than 17 years combined operation experience on more than 12 operating Niro Atomizer spray dryer absorption plants outside the U.S.

The paper describes the European experience with spray dryer absorbers which in many respects is significantly different from the experience in the United States. In Europe more than 70 % of the installed spray dryer absorbers are retrofit installations. In some of these installations the existing electrostatic precipitator is used as dust

collector. In a major part of the retrofit installations, however, the existing electrostatic precipitator is used as pre-collector for fly ash collection. Contrary to the United States, the fabric filters in Europe are of the Pulse-Jet type operating with a high air-to-cloth ratio.

The spray dryer absorbers in Europe operate with low to high sulfur coals with maximum  $\text{SO}_2$  concentrations of  $7.000 \text{ mg/Nm}^3$ . Operating experience has shown that 95 - 97 %  $\text{SO}_2$  removal can consistently be achieved, even on high sulfur coals. The strict European regulations requiring half-hour average for emissions are being achieved with spray dryer absorbers by utilizing a fast-responding feed composition control system.

Operating experience is obtained on base load plants as well as peak load plants with daily start and stop. The operating experience also incorporates the use of high dust SCR systems for  $\text{NO}_x$ -removal upstream of the spray dryer absorber system.

The beneficial effect of the presence of chlorides in the lime slurry was in 1983 originally demonstrated by the US EPA/DOE at the Riverside spray dryer absorber system, Minneapolis. In Europe, coals often contain relatively high chlorine contents and hence operating experience in Europe has shown that a low lime consumption, while maintaining a high stack temperature, can be achieved. Spray dryer absorber plants in Denmark are taking advantage of the chlorine effect by utilizing sea water as make-up in the FGD system.

The paper further makes reference to the first ever spray dryer absorption system in the People's Republic of China. This FGD system will operate on low and high sulfur coal and is as special feature designed to utilize carbide sludge as absorbent.

The paper will finally briefly touch upon the disposal/utilization options being pursued in Europe and China with the dry end product.

### INTRODUCTION

The Spray Dryer Absorption (SDA) process was developed in parallel in the USA and in Europe in the early to mid 70's. The first commercial application of SDA technology for Flue Gas Desulfurization (FGD) on a coal-fired utility boiler was at Northern State Powers Riverside Station in 1980. The pioneers in promoting the SDA-technology, Basin Electric, in North Dakota was the first utility to install a multi-module lime-based SDA system at their Antelope Valley Station, Unit I and II, each with a generating capacity of  $465 \text{ MW}_e$  (1).

Due to the lack of legislation flue gas desulfurization was not applied commercially until the 1980's in Europe. The pioneers in Europe in using the SDA technology were the VKG and EVN utilities which outside Vienna in Austria in 1981 purchased the two first commercial SDA systems with a capacity of  $405 \text{ MW}_e$  and  $352 \text{ MW}_e$  respectively.

Flue gas desulfurization capacity is now applied commercially in many countries in Europe and FGD will continue to grow, as a majority of the countries have committed to reduce their  $\text{SO}_2$  emissions by 30 - 70%.



see Figure 1. The committed reduction in  $\text{SO}_2$  can only be fulfilled by retrofitting a large number of power stations with FGD.

Since the early development of SDA technology, it has been rapidly accepted by the utility industry due to the demonstrated ease of operation, high reliability and low operating costs as compared to the traditional wet scrubbing technology. Today, approximately 11,500  $\text{MW}_e$  of spray dryer absorbers are in operation/under construction on power plants, industrial boilers and incinerators in the following countries (2,3) Austria, Canada, China, Denmark, Finland, Federal Republic of Germany, France, Italy, Japan, Switzerland, Sweden and United States.

Furthermore, SDA technology is under consideration in countries like England, the Soviet Union, Czechoslovakia, Holland, Spain, German Democratic Republic, Hungary, Yugoslavia, Taiwan and Turkey.

Niro Atomizer has been a leader in the development and application of SDA flue gas cleaning technology and has received orders for utility SDA systems representing approximately 8,000  $\text{MW}_e$  (4 - 9).

#### THE NIRO ATOMIZER SPRAY DRYER ABSORPTION PROCESS

Figure 2 is a simplified flow diagram on the SDA process. The reagent for  $\text{SO}_2$  absorption, typically lime, is pumped to the spray dryer absorber, where it is atomized to a cloud of fine droplets. Flue gas containing  $\text{SO}_2$  and fly ash from the boiler enters the spray dryer absorber through gas dispersers. In the spray dryer absorber the atomized droplets get in intimate contact with the flue gas whereby water is evaporated by the heat of the flue gas, while  $\text{SO}_2$  and other acid gases like  $\text{SO}_3$  and  $\text{HCl}$  are simultaneously absorbed. A dry powder is formed as the flue gas is cooled. Finally, the dry powder along with any fly ash is collected and removed from the flue gas in a down-stream filter. The filter can be either a bagfilter or an electrostatic precipitator. Recirculation of the dry end product to the feed system enhances lime utilization (10). The control of the process is simple. The spray dryer absorber outlet temperature is controlled at a low, but safe level above the adiabatic saturation temperature of the flue gas. This provides optimum reagent utilization, while securing a dry, free-flowing product. In another control loop the stack  $\text{SO}_2$  emission is used to control the addition of fresh reagent to the system.

In the Niro Atomizer SDA process, a single rotary atomizer is used to atomize the absorbent into the flue gas. Figure 3 shows an F-800 atomizer and the abrasion resistant KN-wheel. The use of a single rotary atomizer in each spray dryer absorber makes the process easy to control and avoids operational problems. The high speed rotary atomizer is designed for long life and is outfitted with an abrasion resistant wheel. The wheel consists of a wheel body, made of stainless steel or more corrosion resistant alloy and inserts protruding into the wheel body to form a layer of slurry for protection of the inside of the wheel body when the wheel is rotating. The bottom plate and the inserts are made of abrasion resistant material, e.g. Silicon carbide.

Key features of the SDA process are listed in Figure 4.

## PLANT DESCRIPTIONS

The European spray dryer absorptions systems differ in a number of ways from the spray dryer absorptions system installed in the US. Figure 5 shows some key design data.

In Europe the legislation does not require installation of a spare module. Thus the European SDA systems operate at full load with no spare module. The Duernrohr plant however has an additional overload handling capacity by virtue of the dimensions of the 3 modules, each of which can handle 40 % of full load capacity. A bypass around the absorbers is not used in any of the installations either.

The use of a precollector for fly ash collection is a common design feature for the major part of the European SDA plants, where waste product disposal is a major concern. In Europe there is a well established market for fly ash utilization, thus precollection yields a saleable product and furthermore decreases the volume of FGD waste for further utilization/disposal. The precollector is normally a one-field electrostatic precipitator, however in retrofit situations the existing electrostatic precipitator is often being used as precollector.

The use of electrostatic precipitators as main filters is very popular in Europe and thus 1/3 of the SDA systems on order are equipped with 3-5 field electrostatic precipitators. The rest of the SDA systems incorporate high ratio pulse-jet fabric filters for final dust collection. From Figure 5 it appears that the Walheim power station is equipped with two pulse jet filters, with a capacity of 105 MW and 155 MW respectively, which today are among the worlds largest of the kind. This however will be surpassed by the two 350 MW pulse jet filters at the Midtkraft power station going on stream in early 1989 and 1990. The first SDA system in the People's Republic of China will also apply a pulse-jet fabric filter as dust collector.

As European legislation for SO<sub>2</sub> reduction does not apply to flue gas desulfurization on new power stations only, 2/3 of the SDA systems are retrofit applications. It should be pointed out that at 2 power stations, Aroskraft and VW, the existing electrostatic precipitators have been used as main filter for the SDA system and at these plants no precollector has been installed for fly ash collection.

In Europe stack temperature requirements vary from 65 - 85°C. The stack temperature requirement can often be met by adjusting the spray dryer absorber outlet temperature. In plants where a tail end SCR De-NO<sub>x</sub> system is installed, no reheat is required of course.

Figure 6 shows that at some SDA installations SCR systems for NO<sub>x</sub> removal are installed upstream the spray dryer absorber, whereas 3 spray dryer absorber systems have a tail end low dust SCR system for DeNO<sub>x</sub>.

In the US ball mill slakers are typically used for lime slaking in the SDA systems. In Europe the majority of the SDA/FGD systems are equipped with detention type tank slakers, except the Fynsvaerket power station which will employ tower mill slakers. The simple tank slaker can be used for pulverized quick lime. The tank slaker at Midtkraft is an attrition type slaker which allows slaking of 2-13 mm pebble lime. The use of

tower mills for slaking was originally proposed by Dharma et al. (11). Recent investigations by Niro Atomizer have shown that tower mill slaking for some lime types can increase the reactivity of the slaked lime. In the People's Republic of China, highly reactive quick limes are not readily available. The SDA system incorporates a tank slaker with extended slaking time for slaking of lime with approximately 60% active CaO. This plant is also designed for using carbide lime with an active lime content of 72 - 83 %. It is of great benefit to the Chinese to utilize a carbide waste as this is more economical and solves an environmental problem (no disposal of carbide sludge).

In Europe SDA systems are supplied as turn key systems. In China however the scope of supply is rather limited as the Chinese have the goal to be able to do as much as possible themselves. In the first SDA system in Shenyang, Liaoning Province, Niro Atomizer/Flakt Denmark are supplying key components such as atomizer, gas disperser, product conditioner, double flap valves, all instrumentation and the control system. Also basic engineering and technical assistance is supplied to secure a successful start-up and operation of the plant. Detailed engineering for the balance of the plant is being done by China Aeronautical Project and Design Institute and Beijing Municipal Institute of Labour Protection (BMILP).

The process design of the European SDA systems has to take into account a number of factors. The use of spot market coal is a common feature for many of the European power stations, see Figure 7. An example of the various coals that were burned at the Aroskraft power station during one year is shown in Figure 8. Accordingly, the lime storage and slaking equipment are designed to handle the large variation in lime consumption. Most of the coals burned in Europe have a relatively high chlorine content. The presence of chlorine reduces the lime consumption for a given SO<sub>2</sub>-removal considerably. This was demonstrated by the US EPA/DOE already in 1983 (12). Furthermore, the content of chloride in the end product determines the delta T to adiabatic saturation at which the plant safely can be operated. With chloride concentrations in the end product up to 2 %, the SDA system can be operated at 10°C whereas a content of 5 % chloride requires an approach temperature of 25°C. In Denmark, the Midtkraft and Fynsværket SDA systems are designed to partly utilize seawater as make-up water in the feed system. Thereby the chloride effect can be optimized and in case of Midtkraft the required stack temperature of 20°C above saturation can be met without re-heat.

A special design feature of the European SDA systems is the use of a fast-response feed system. A fast-response system is required to be able to meet the stringent half-hour SO<sub>2</sub>-emission values, which are in force in f. inst. West Germany. The day-to-day variations in sulfur content also require a fast-response system to meet the half-hour average.

There is no uniform legislation in Europe with respect to required SO<sub>2</sub>-removal. In West Germany a minimum SO<sub>2</sub> removal of 85 %, however a maximum emission value of 400 mg/Nm<sup>3</sup> is required. In Austria the same outlet emission of 400 mg/Nm<sup>3</sup> SO<sub>2</sub> is applied plus a further requirement of minimum 90 % SO<sub>2</sub>-removal regardless of inlet SO<sub>2</sub> concentration. In Sweden the requirement is based on a yearly sulfur removal corresponding to an average of 80 % removal. Denmark utilizes a bubble concept for SO<sub>2</sub> emissions. This allows the cooperation of Danish power stations Elsam and



Elkraft to optimize the FGD systems such that  $\text{SO}_2$  is removed at minimum cost per ton of  $\text{SO}_2$  removed (13).

In the People's Republic of China no legislation for  $\text{SO}_2$  control has been promulgated yet. However China is on its way to industrialization and 70 % of the total fuel consumption is coal. The total  $\text{SO}_2$  emission was 15 million tons in 1982 and with no control it is expected to reach 32 million tons in the year 2000. Already today 50 % of all rain samples in China are acid ( $\text{pH} < 5.6$ ). Facing this environmental problem a search for proper control technology has become a public interest. In 1985, authorized by the State Science and Technology Commission of China (SSTCC) and national EPA and further supported by the State Economy Commission of China (SECC) and Ministry of Aviation Industry, the spray dryer absorption technology was selected to be the first demonstration plant, because of its simplicity and cost effectiveness. The SDA plant being located in Shenyang is designed for 70 % and 90 % removal of  $\text{SO}_2$  both for low and high sulfur coals.

As it appears from Figure 7 the European SDA systems are designed for low and high sulfur coal with the inlet  $\text{SO}_2$  concentrations from 2.000 to 7.000  $\text{mg/Nm}^3$  (700 - 2.500 ppm). Required  $\text{SO}_2$  removal is as high as 95 % for low sulfur coal and 97 % for high sulfur coal. It should be emphasized that these high removal efficiencies can also be achieved with an electrostatic precipitator as dust collector.

## OPERATING AND TEST RESULTS

### GENERAL EXPERIENCE

By now 12 out of 17 European Niro Atomizer SDA systems are in operation. 2/3 of these plants have gone through formal performance and acceptance testing and have met all guarantees. Test results from 8 of the plants are shown in Figure 9. The test results from Aroskraft and from VW clearly show the beneficial effects of the presence of chloride in the feed slurry. Also it is seen that the operating delta T adsat is adjusted according to chloride concentration. When chlorides are present, low stoichiometries can be obtained even at high delta T's. The test results from Duernrohr and Walheim, Erlangen and Braunschweig demonstrate that high removal efficiencies of  $\text{SO}_2$  easily can be achieved on low sulfur coal with a reasonably low stoichiometry. The results from the SDA system at Salzburg are very interesting. This is the first time it commercially has been proven, that 97 %  $\text{SO}_2$  removal is feasible on high sulfur coal. It should be mentioned that 97 % removal efficiency is as an half-hour average. The  $\text{SO}_2$  concentration of approx. 7.000  $\text{mg}$  (2.500 ppm) in the flue gas corresponds to 3.5 - 4 % sulfur in normal Eastern US coals.

### THE AROSKRAFT POWER STATION

At the Aroskraft power station, the operating experience with the SDA system has generally been very good. At Aroskraft, a unique problem was encountered in the initial phase of operation. Extremely abrasive fly ash particles were wearing out the nozzles in the atomizer wheel so fast that the nozzle had to be turned every 100-200 hours. The boiler had previously been converted from oil to coal and it was the suspicion that



there was not sufficient of residence time to melt the fly ash and thereby making it less abrasive. The problem was solved by directing the product from the spray dryer absorber directly to disposal instead of using it for recycle. The conditioning effect of the spray dryer absorber on the existing electrostatic precipitator has clearly been demonstrated. Outlet emissions are lower than before the installation of the FGD system and now any coal can be burned without dust emission problems. Previously, low sulfur coal from Australia and Colombia could not be burned on this plant without exceeding the dust emission limits. After installation of the SDA system emissions are low regardless of coal type. During the first year of operation, experience was obtained with two different types of lime. Due to differences in reactivity, a difference of 20 % in the lime consumption for a given  $\text{SO}_2$ -removal was measured.

#### THE DUERNROHR POWER STATION

At the Duernrohr power station, Figure 10, the SDA system has shown very good performance (14). During the initial operating period problems with the feed preparation area required more than average maintenance. Deposits in some of the spray dryer absorbers and on the emitting electrodes in the first field of the electrostatic precipitator were once experienced as a problem. This problem was solved by adjusting the gas split between gas dispersers and by increasing the hammer rapping frequency on the electrostatic precipitator. Further experience from Duernrohr shows that overloading of the SDA modules and the load following capability is very good.  $\text{SO}_2$ -removal efficiencies up to 95 % can be achieved during normal operation. At the EVN utility, 10 different types of coals from all over the world were tested over a period of 2 months. Good operating experience was achieved with the SDA system and again it was proven that the performance of the electrostatic precipitator is unaffected by the type of coal being burned. These tests further verified that up to 25 % of lime can be saved by the presence of chloride in the coal. The mercury removal capability of the spray dryer absorber system was investigated. Figure 11 shows that approx. 90 % of the mercury present in the gas phase was removed. This is a significant result compared with mercury removal in wet scrubbers which are reported to be 40 - 50 % (15).

Also at Duernrohr, different lime qualities were tested - Austrian, Polish and Yugoslavian lime. It was demonstrated that Polish and Yugoslavian lime due to the lower reactivity led to much higher lime consumption than when running the plant with Austrian quick lime.

The Duernrohr power station is equipped with a high dust SCR system upstream the spray dryer absorbers. There have been no operational problems due to ammonia carry-over from the SCR system into the FGD system.

#### THE POWER STATION IN SALZBURG

The expectations and requirements to removal efficiency, availability and ease of operation were achieved. During the first year of operation,  $\text{SO}_2$  concentrations were fluctuating between 4.000 and 7.000  $\text{mg}/\text{Nm}^3$  and the corresponding  $\text{SO}_2$ -removal efficiency was 95 - 97.1 % to meet the required outlet emission of 200  $\text{mg}/\text{Nm}^3$ .

## THE WALHEIM POWER STATION

The Walheim SDA system has been operating quite well. Some mechanical problems were experienced with powder feeding and transport systems as well with tank agitators. Fluctuations in the amount of excess lime in the recyle slurry combined with a chloride concentration of 20.000 - 40.000 ppm has led to pit-corrosion in the tanks, especially in areas with deposit build-up. The tanks are made of mild steel and various solutions are currently being investigated to solve this problem. The dust emission from the pulse-jet filter has been less than 10 mg/Nm<sup>3</sup>. Some problems have been experienced with deposits on the bags. Furthermore, problems with broken clamps and corrosion of galvanized parts have been experienced.

The Walheim power station is a peak load plant. The SDA system hence follows the swinging load with daily/weekly start and stops.

The SDA has been operating so effectively that no or very little excess lime was found in the end product. As a certain lime content (4 - 8 %) is required for disposal purposes the spray dryer absorber outlet temperature was increased a few degrees, which increased the excess lime content to the required level.

## AVAILABILITY

The availability of the European SDA systems has generally been very good. Figure 12 shows availability data for the Duernrohr, Aroskraft, Salzburg and Walheim power stations. At Duernrohr and Aroskraft the availability has virtually been 100 %. At the SDA system in Salzburg an availability of 97.0 % was recorded during the first year of operation. The lost availability was due to a number of minor mechanical problems, which now have been correct. At the Walheim power station the unavailability has been caused by initial mechanical problems which now have been solved and recently availability has typically been in the high 90's.

## END PRODUCT DISPOSAL AND UTILIZATION

In Europe disposal and utilization of various FGD end products is being discussed intensively between the countries in order to come up with common guide lines (16)(18).

The dry, free-flowing product from the SDA process can be considered as a new industrial raw material (19). The most frequent way of disposing off SDA end products is in stabilized landfills. This is the overall dominating way of disposal in the United States and an impressive example of this disposal technique is shown in Figure 13. The US practice of stabilization has been adopted in Europe. In Denmark f. inst. the Danish utility Midtkraft has planned a land reclamation project at Glatved as a backup disposal possibility for the coming SDA end product. The similarity between the US and the Danish project can be seen by comparing Figure 13 and 14. In the Danish project it is an old gravel pit that is being restored while in the US example it is an old strip mine that is

being restored, in both cases in order to re-create a spoiled water front. This landscaping technique is an environmentally acceptable way of utilizing SDA products (17).

In Europe utilization of the FGD end product has a high priority due to the limited space for disposal by stabilized landfilling. As more and more plants come on line, the SDA product becomes available and promotes intensified research into alternative uses of the SDA product. Unrecognized technical properties have now come into light making the dry product a new, though unconventional raw material in much the same way as coal fly ash were many years ago.

In Austria the VKG and EVN utilities are investigating various utilization options for the SDA end product. One option is to use the end product in construction materials such as concrete, sand-lime bricks, synthetic gravel etc. Other options being considered are substitution of construction materials in underground works and as ceiling layers for dumps. Full scale tests have verified the usefulness for ceiling layers. Furthermore EVN and VKG consider to convert the SDA end product into technical Anhydrite which then can be used as a cement retarder. The last option being pursued is the use of the dry end product in the Müller-Kühne method for producing sulfuric acid and cement klinker.

In W. Germany the option of making technical Anhydrite (Cement-Gypsum) is being pursued by a German district heating power station (20). This utilization option however has the disadvantage that it competes with FGD gypsum.

In W. Germany SDA end products are being shipped to limestone quarries where they as stabilized landfills are used to backfill the quarry (21).

At the Aroskraft power station in Sweden, a light-weight concrete is produced from the FGD end product. The product is called "Cefill" and is primarily being applied for membranes in old waste disposal sites.

In the People's Republic of China the end product from the first spray dryer absorber system will be used as test material for production of sulfuric acid and cement klinker in the Müller-Kühne process.

Figure 15 summarized the disposal and utilization options being pursued in Europe and in the People's Republic of China.

#### NEW DEVELOPMENTS OF THE APPLICATION OF SDA TECHNOLOGY

Several countries in both Eastern and Western Europe are interested in combining boiler limestone injection (LIMB) with spray dryer absorption. Niro Atomizer has run pilot tests with several different LIMB materials. The test shows that activation of the LIMB materials results in excellent  $\text{SO}_2$  removal in the spray dryer absorber. Under optimum conditions with respect to limestone injection into the boiler and LIMB activation an overall  $\text{SO}_2$  removal of 90 % from  $7.000 \text{ mg/Nm}^3$  has been achieved with a limestone stoichiometry of only 1.5. In these tests an electrostatic precipitator was used as dust collector.

In Europe regenerative systems are seriously being considered for FGD as this partly solves problems with disposal of FGD waste.

The SDA process has the flexibility that various absorbents can be used without radical changes in the basic FGD system (Figure 16). A regenerative SDA system is currently being developed in Denmark with MgO as absorbent. The development program is supported by Danish utility companies. Pilot plant tests have shown that 90 % SO<sub>2</sub> removal on 2 % sulfur coal can be achieved with a reasonable MgO stoichiometry. The produced magnesium sulfite has been regenerated in a high temperature fluid bed without problems with de-activation of the material, on the contrary a more reactive material is achieved after regeneration compared with the fresh MgO make-up.

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| Country        | 1983 Sulfur<br>Emission<br>Tonnes x 10 <sup>3</sup> | Reduction<br>Planned (Basis 1980) |      |
|----------------|-----------------------------------------------------|-----------------------------------|------|
|                |                                                     | %                                 | Year |
| Austria        | 147                                                 | 70                                | 1995 |
| Belgium        | 306                                                 | 50                                | 1995 |
| Czechoslovakia | 1625                                                | 30                                | 1993 |
| Denmark        | 151                                                 | 50                                | 1995 |
| E. Germany     | 2000                                                | 30                                | 1993 |
| Finland        | 180                                                 | 50                                | 1993 |
| France         | 1015                                                | 50                                | 1990 |
| Hungary        | 825                                                 | 30                                | 1993 |
| Ireland        | 70                                                  | -                                 | -    |
| Italy          | 1900                                                | 30                                | 1993 |
| Luxembourg     | 14                                                  | 58                                | 1990 |
| Netherlands    | 170                                                 | 50                                | 1995 |
| Norway         | 50                                                  | 50                                | 1994 |
| Poland         | 2050                                                | -                                 | -    |
| Soviet Union   | 6000                                                | 30                                | 1993 |
| Spain          | 1638                                                | -                                 | -    |
| Sweden         | 165                                                 | 68                                | 1995 |
| Switzerland    | 43                                                  | 57                                | 1995 |
| United Kingdom | 1845                                                | 30                                | 1999 |
| West Germany   | 1375                                                | 65                                | 1993 |

Fig. 1 SO<sub>2</sub> Emissions and Planned Reduction in Europe.

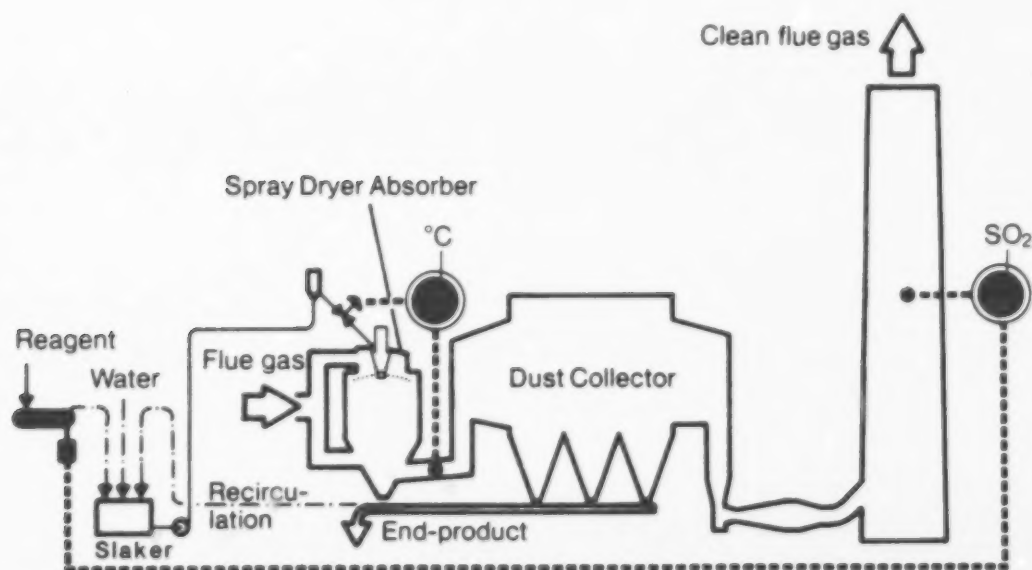


Fig. 2 Spray Dryer Absorber Process

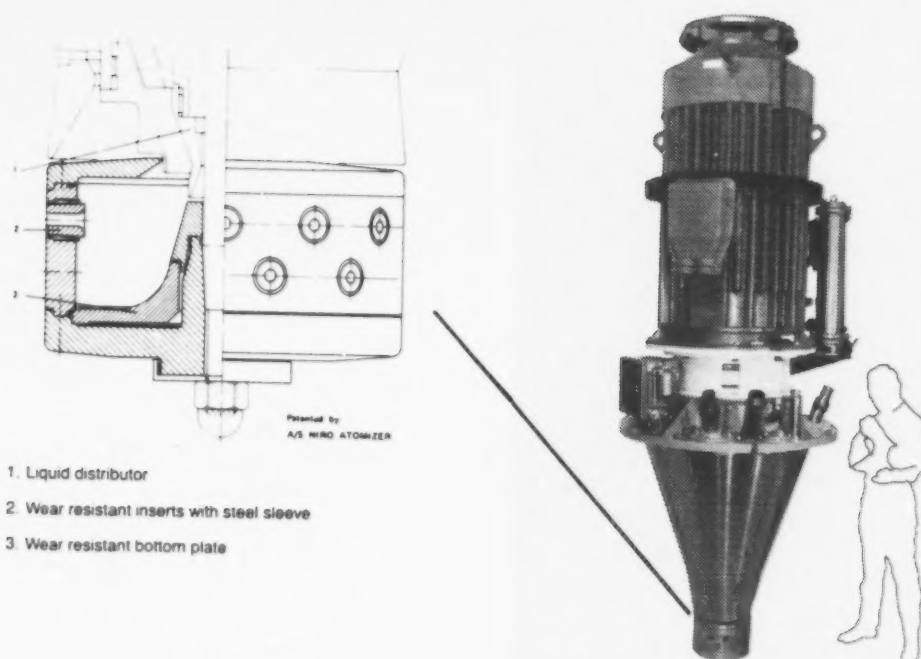


Fig. 3 Rotary atomizer and abrasion resistant atomizer wheel design

- \* Simple process, known for 50 years
- \* Single Rotary Atomizer
- \* No corrosion - normal carbon steel used as construction material
- \* Electrostatic precipitator or bagfilter as dust collector
- \* Dry end product - no waste water
- \* Low power consumption
- \* Applicable to low and high sulfur coal
- \* 90 - 95 %  $\text{SO}_2$  removal
- \* Flexible. Various absorbents can be used and various end products can be produced
- \* Product recycling exchanges reagent utilization

Fig. 4 Key features of the Niro Atomizer FGD process



| PLANT          | COUNTRY*) | SIZE<br>MWe | RETRO-<br>FIT | NO. OF<br>MODULES | PRE-<br>COLL. | MAIN<br>FILTER | RE-<br>HEAT | START-<br>UP |
|----------------|-----------|-------------|---------------|-------------------|---------------|----------------|-------------|--------------|
| AROSKRAFT      | S         | 420 e+th    | YES           | 1                 |               | ESP            | NO          | 10/86        |
| BRESCIA        | I         | 60          |               | 1                 | YES           | FF             |             | 8/87         |
| BRAUNSCWHEIG   | D         | 170 e+th    | YES           | 1                 | YES           | FF             | NO          | 9/86         |
| BMILP          | CHINA     | 6           | YES           | 1                 |               | FF             | NO          | 11/88        |
| DUERNROHR, VKG | A         | 405         |               | 3                 | YES           | ESP            | TO 85 °C ** | 5/85         |
| DUERNROHR, EVN | A         | 350         |               | 3                 | YES           | ESP            | TO 85 °C ** | 9/86         |
| ENDSOLF        | D         | 410         | YES           | 3                 | YES           | ESP            | NO          | 4/88         |
| ERLANGEN       | D         | 88          | YES           | 1                 | YES           | FF             | NO          | 8/87         |
| FYNSVAERKET    | DK        | 350         |               | 2                 | YES           | FF             | NO          | 7/91         |
| HANNOVER       | D         | 260         |               | 2                 |               | ESP            | NO          | 12/89        |
| HUELS          | D         | 70          | YES           | 1                 | YES           | FF             | NO          | 9/88         |
| MIDTKRAFT      | DK        | 350         | YES           | 2                 | YES           | FF             | NO          | 1/89         |
| MIDTKRAFT      | DK        | 350         | YES           | 2                 | YES           | FF             | NO          | 1/90         |
| MAINKRAFTWERKE | D         | 150 e+th    | YES           | 1                 | YES           | FF             | NO          | 7/88         |
| SALZBURG       | A         |             | YES           | 1                 |               | FF             | NO          | 10/87        |
| VW             | D         | 260         | YES           | 2                 |               | ESP            | TO 80 °C    | 8/87         |
| WALHEIM        | D         | 105 + 155   | YES           | 2                 | YES           | FF             | TO 77 °C    | 6/87         |

\*) A = Austria D = W. Germany DK = Denmark  
I = Italy S = Sweden

\*\* Installed but not necessary

Fig. 5 Design data for Niro Atomizer European and Chinese FGD system

| PLANT          | HIGH DUST SCR    | TAIL END LOW DUST SCR |
|----------------|------------------|-----------------------|
| BRAUNSCHWEIG   |                  | IN DESIGN/ERECTION    |
| DUERNROHR, VKG | IN OPERATION     |                       |
| DUERNROHR, EVN | IN OPERATION     |                       |
| ENDSOLF        |                  | IN DESIGN/ERECTION    |
| FYNSVAERKET    | BEING CONSIDERED |                       |
| MIDTKRAFT      | BEING CONSIDERED | BEING CONSIDERED      |
| WALHEIM        | IN OPERATION     |                       |
| HANNOVER       |                  | IN OPERATION          |

Fig. 6 DeNOx-installations in connection with European dry FGD plants

| PLANT          | COUNTRY*) |           | COAL      |         | FLUE GAS                                      |                |                                       | FGD SYSTEM |                  |
|----------------|-----------|-----------|-----------|---------|-----------------------------------------------|----------------|---------------------------------------|------------|------------------|
|                | ORIGIN    |           | % S       | % CL    | FLOW<br>Nm <sup>3</sup> /hr x 10 <sup>3</sup> | TEMP.<br>DEG C | SO <sub>2</sub><br>mg/NM <sup>3</sup> | % REM.     | DELTA T<br>DEG C |
| AROSKRAFT      | S         | VARIOUS   | 0.3 - 1.5 | 0 - 0.3 | 550                                           | 120            | 1965                                  | 80.0       | 10               |
| BRESCIA        | I         | VARIOUS   | 0.5 - 3.0 | -       | 220                                           | 125            | 6800                                  | 93.3       | 10               |
| BRAUNSCWHEIG   | D         | VARIOUS   | 0.5 - 1.4 | 0 - 0.4 | 210                                           | 130            | 1920                                  | 95.0       | 15               |
| BMILP          | CHINA     | N. CHINA  | 1.5 - 3.0 | 0       | 50                                            | 160            | 5700                                  | 90.0       | 15               |
| DUERNROHR, VKG | A         | POLAND    | 0.6 - 1.0 | 0 - 0.2 | 1400                                          | 125            | 2300                                  | MIN. 90    | 15               |
| DUERNROHR, EVN | A         | P/VARIOUS | 0.6 - 1.0 | 0 - 0.2 | 1100                                          | 134            | 2300                                  | MIN. 90    | 15               |
| ENSDORF        | D         | SAAR      | 0.9       | 0.2     | 1780                                          | 130            | 3000                                  | 95.0       | 10               |
| ERLANGEN       | D         | RUHR/SAAR | 1.2       | 0.15    | 108                                           | 134            | 2390                                  | 92.0       | 30               |
| FYNSVAERKET    | DK        | VARIOUS   | UP TO 2.5 | 0.1     | 1340                                          | 125            | 4000                                  | 96.5       | 10               |
| HANNOVER       | D         | VARIOUS   | 0.4 - 1.4 | 0.1     | 980                                           | 130            | 5000                                  | 94.0       | 10               |
| HUELS          | D         | RUHR      | 1.2       | <0.1    | 286                                           | 130            | 2400                                  | 95.0       | 10               |
| MIDTKRAFT      | DK        | VARIOUS   | UP TO 2.5 | 0.1     | 1240                                          | 125            | 3000                                  | 93.0       | 20               |
| MIDTKRAFT      | DK        | VARIOUS   | UP TO 2.5 | 0.1     | 1240                                          | 125            | 4000                                  | 92.0       | 20               |
| MAINKRAFTWERKE | D         | RUHR      | 1.2       | <0.1    | 390                                           | 120            | 2500                                  | 92.0       | 20               |
| SALZBURG       | A         | LIGNITE   | 2.0       | 0       | 115                                           | 160            | 7000                                  | MIN. 90    | 25               |
| VW             | D         | VARIOUS   | 0.4 - 1.3 | 0.1     | 980                                           | 130            | 2400                                  | 90.0       | 10               |
| WALHEIM        | D         | VARIOUS   | 0.7 - 1.3 | <0.3    | 980                                           | 125            | 2515                                  | 90.0       | 10               |

\*) A = AUSTRIA    D = W. GERMANY  
 DK = DENMARK    I = ITALY  
 S = SWEDEN

Fig. 7 Process design data for Niro Atomizer European and Chinese FGD System

| ORIGIN    | % S  | % CL  |
|-----------|------|-------|
| USSR      | 0.38 | 0.01  |
| POLAND    | 0.57 | 0.15  |
| USA       | 1.16 | <0.01 |
| COLOMBIA  | 0.55 | 0.04  |
| AUSTRALIA | 0.38 | 0.03  |
|           | 0.56 | 0.02  |
| ENGLAND   | 1.49 | 0.02  |
| VENEZUELA | 0.47 | 0.01  |

Fig. 8 Spot market coal burnt at Aroskraft Power station 1987/1988

|                | COAL                   |      | TEMP.<br>DEG.C | FLUEGAS                               |                                             | T<br>DEG.C | FGD SYSTEM                 |                  |                     | FIL-<br>TER |
|----------------|------------------------|------|----------------|---------------------------------------|---------------------------------------------|------------|----------------------------|------------------|---------------------|-------------|
|                | % S                    | % Cl |                | SO <sub>2</sub><br>mg/Nm <sup>3</sup> | FLOW<br>Nm <sup>3</sup> /hrx10 <sup>3</sup> |            | SO <sub>2</sub> -REM.<br>% | STOICH.<br>RATIO | % Cl IN<br>ENDPROD. |             |
| AROSKRAFT      | 0.6                    | 0.2  | 118            | 1410                                  | 542                                         | 20         | 82.4                       | 1.02             | 2.5                 | ESP         |
|                | 1.2                    | 0.0  | 117            | 2486                                  | 498                                         | 9          | 80.0                       | 1.23             | 0.0                 | ESP         |
| DUERNROHR, VKG | 0.6                    | 0.1  | 136            | 1356                                  | 1369                                        | 15         | 91.2                       | 1.10             | 1.0                 | ESP         |
|                | 0.6                    | 0.1  | 140            | 1356                                  | 1070                                        | 18         | 91.2                       | 1.14             | 1.0                 | ESP         |
| SALZBURG       | 2.0                    | 0.0  | 180            | 6857                                  | 115                                         | 10         | 97.1                       | 1.60             | 0.0                 | FF          |
| VW             | 1.0                    | 0.1  | 131            | 1560                                  | 465                                         | 10         | 90.1                       | 1.13             | 0.8                 | ESP         |
| ERLANGEN       | MIXED<br>OIL &<br>COAL |      | 122            | 756                                   | 107                                         | 33         | 94.1                       | 1.20             | 2.6                 | FF          |
| WALHEIM        | 0.7-1.3                | 0.2  | 126            | 2000                                  | 980                                         | 18         | 95.0                       | 1.1              | 2.6                 | FF          |
|                |                        |      | 130            | 1500                                  |                                             | 17         | 95.0                       |                  |                     |             |
| BRAUNSCHWEIG   | 1.1                    | 0.1  | 150            | 1650                                  | 220                                         | 15         | 95.0                       | 1.08             | 1.5                 | FF          |

Fig. 9 Test Results from Niro Atomizer European FGD systems.



Fig. 10 Duernrohr Power Station

| TEST | LOCATION | $\mu\text{g}/\text{Nm}^3$ Hg | % Removal |
|------|----------|------------------------------|-----------|
| 1    | SDA in   | 2.74                         | 88        |
|      | ESP out  | 0.32                         |           |
| 2    | SDA in   | 2.62                         | 89        |
|      | ESP out  | 0.29                         |           |

Fig. 11 Mercury removal test results from Duernrohr power station.

| PLANT          | AVAILABILITY | PERIOD          |
|----------------|--------------|-----------------|
| AROSKRAFT      | 100          | SINCE OCT. 1986 |
| DUERNROHR, VKG | 100          | SINCE 1985      |
| DUERNROHR, EVN | 100          | SINCE SEP. 1986 |
| WALHEIM        | 95.7         | SINCE JAN. 1988 |
| SALZBURG       | 97.0         | SINCE SEP. 1987 |

Fig. 12 SDA Availability Data



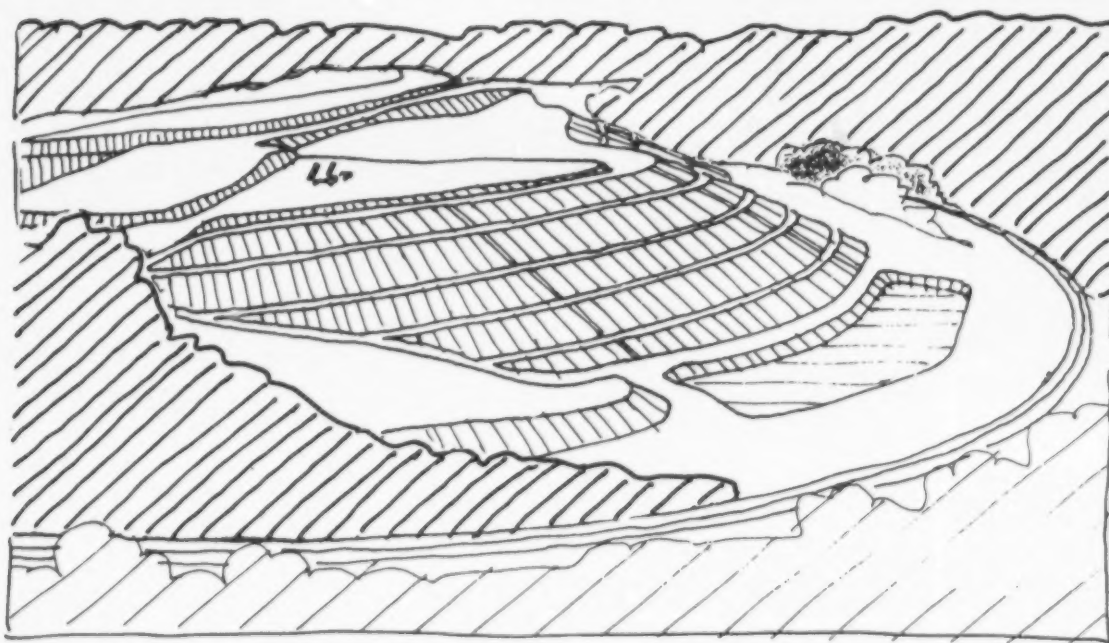


Fig. 13 Re-establishment of old strip mining area using combustion residues.

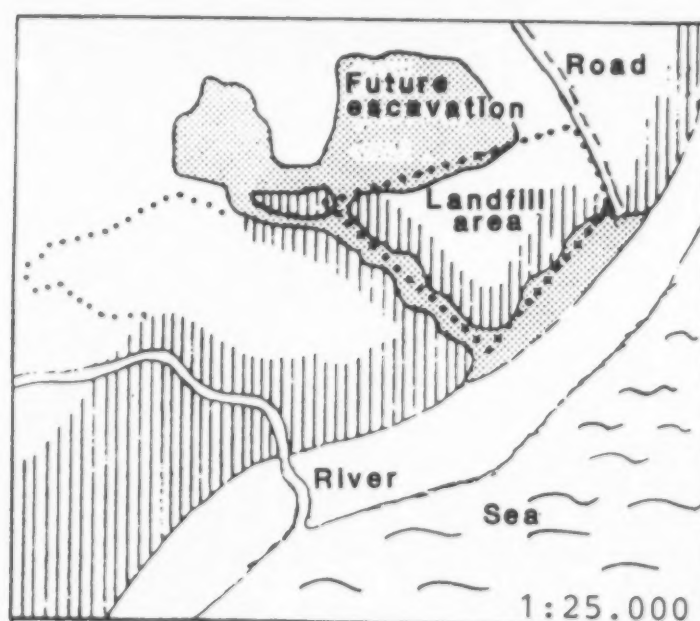


Fig. 14 Re-establishment of part of limestone quarry using combustion residues (Studstrupvaerket, Glatved, DK).

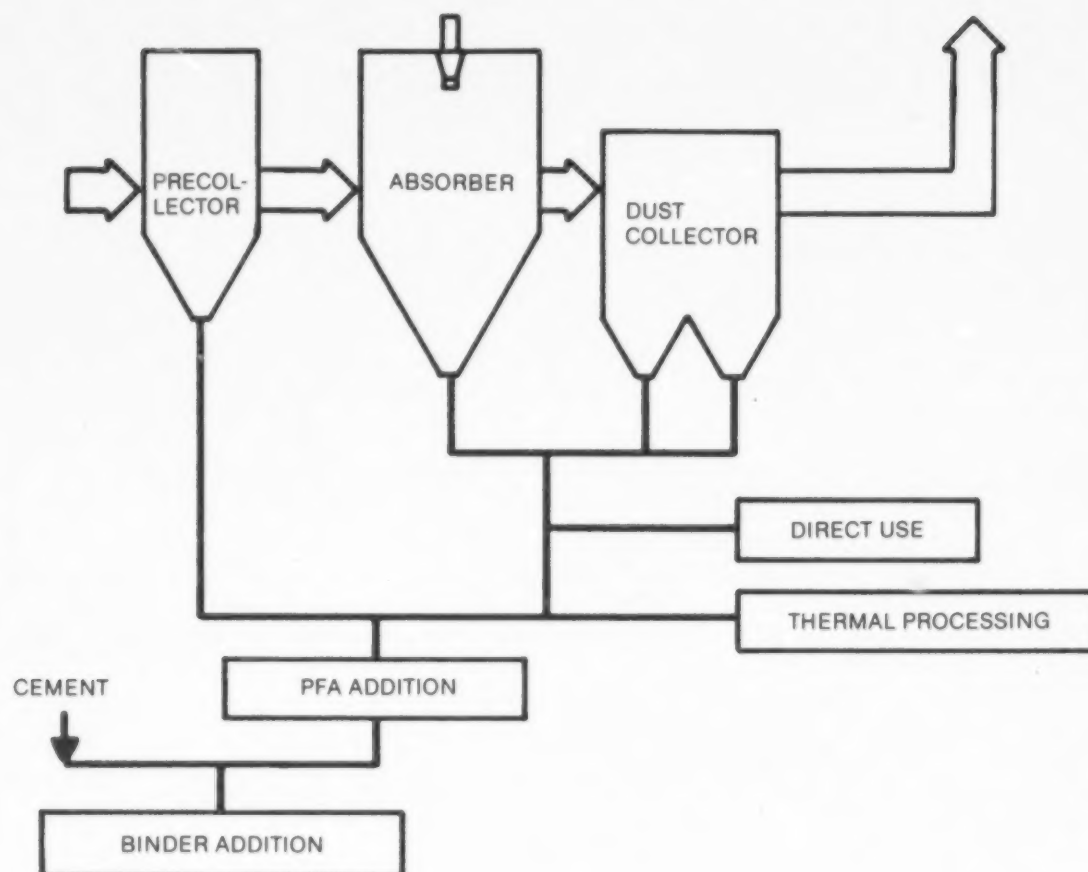


Fig. 15 European/Chinese disposal and utilization options for SDA end products.

#### PROCESSING

##### Direct use

##### Thermal Processing

Blended with Fly Ash  
and for some application  
further binder addition

#### APPLICATION

Soil stabilized landfill

Additive to:

- \* cellular and plain concrete
- \* fiber boards
- \* sand - lime bricks, etc.

Technical Anhydrite  
Cement/Sulfuric Acid

Landfill/Landscaping  
Marine Landreclamation/Ocean Reefs  
Gravel/Embankments  
Grouts for

- \* Pipeline trenches
- \* Membranes
- \* Gravity filling
- \* Road Basis

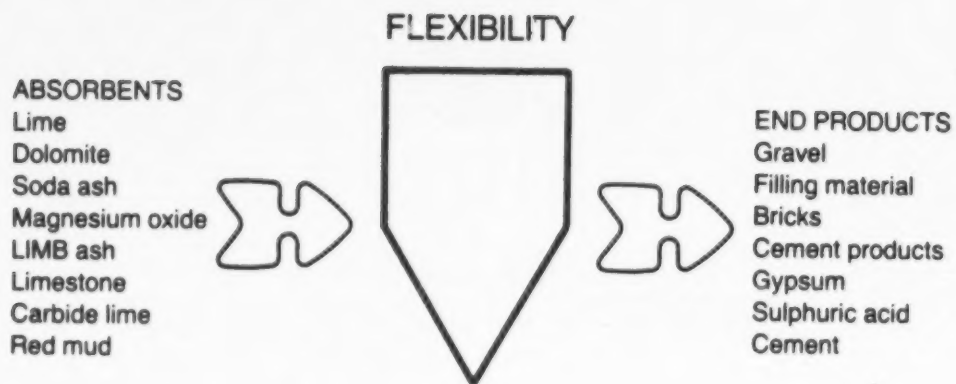


**Fig. 16**

## RESULTS FROM EPRI HSTC HIGH SULFUR SPRAY DRYER PILOT TESTS

by: Gary Blythe and Larry Lepovitz  
Radian Corporation  
Austin, Texas 78720-1088

Richard Rhudy  
Electric Power Research Institute  
Palo Alto, California 94303

### ABSTRACT

This paper presents results from pilot-scale tests conducted to evaluate spray dryer FGD technology for new and retrofit high-sulfur-coal applications. The tests are being conducted on a 4-MW spray dryer FGD system at the EPRI High Sulfur Test Center. Both fabric filter and electrostatic precipitator (ESP) particulate control devices have been tested with the spray dryer.

Most of the SO<sub>2</sub> removal data are for operation with the fabric filter particulate collector. For a spray dryer inlet temperature of 300°F, and an inlet SO<sub>2</sub> level of 2500 ppmv, 90% overall SO<sub>2</sub> removal was achieved at a calcium-to-sulfur ratio of 1.3 to 1.6, depending on the lime source used. For one lime source, raising the spray dryer inlet temperature to 350°F decreased the calcium-to-sulfur ratio requirement to achieve 90% overall removal from 1.6 to about 1.2. For an inlet SO<sub>2</sub> level of 2000 ppmv, a 300°F inlet temperature, and the most reactive of the limes tested, the calcium-to-sulfur ratio required to achieve 90% overall removal was lowered to 1.0. In two tests with the ESP rather than fabric filter used as a particulate control device, overall SO<sub>2</sub> removal levels were reduced by 3 to 6 percentage points relative to that achieved at similar conditions with the fabric filter.

The fabric filter was found to maintain particulate emissions well below the current New Source Performance Standards (NSPS) levels. After approximately 8000 hours of operation, and about 60 start-ups and shutdowns, pressure drop performance and bag residual dustcake weights remained at acceptable levels. There were some adverse effects of the spray dryer on fabric filter corrosion tendencies. Compartment wall corrosion was encountered, and appears to be best addressed by improved insulation practices and perhaps application of protective coatings. Failures of 300-series stainless steel bag clamps were also encountered, because of the presence of chloride from the coal. Such failures would be best avoided by using chloride resistant materials for bag clamps in spray dryer FGD systems for coal with an appreciable chloride content. The ESP was able to maintain particulate emissions below the current NSPS levels when the spray dryer was in service upstream, with 4 fields operating and an SCA of about 400 ft<sup>2</sup>/kacfm. With fewer fields and a lower SCA, current NSPS levels were exceeded, apparently due to scouring of collected particles from the plate surfaces.



## INTRODUCTION

There are 15 utility spray dryer flue gas desulfurization (FGD) systems in operation in the U.S., treating flue gas from over 5000 MW of generating capacity. Of these, 12 have fabric filter particulate control devices, and 3 have ESPs. All of these systems are installed on boilers that fire low- to medium-sulfur-content coal. A short-term demonstration co-funded by EPRI and the U.S. EPA in 1983 documented that a utility spray dryer/fabric filter FGD system was capable of achieving 90%  $\text{SO}_2$  removal when treating flue gas from a high-sulfur (3.2%) coal (1). However, there has been no commercial application of spray dryer FGD technology to a utility boiler that regularly fires a high-sulfur coal.

Pilot-scale testing of spray dryer/fabric filter FGD technology conducted by EPRI at their Arapahoe test facility, at conditions which simulated high-sulfur-coal firing, have similarly shown that overall  $\text{SO}_2$  removal levels of greater than 90% are achievable. These tests were conducted by spiking  $\text{SO}_2$  into flue gas from a low-sulfur-coal-fired source. However, there are concerns that differences between low-sulfur coal and actual high-sulfur-coal firing (such as ash characteristics and flue gas  $\text{SO}_3$  content) may influence spiked test results. An example of the differences between low-sulfur and high-sulfur coal is the effect on a fabric filter particulate control device. Reverse-gas fabric filters installed in high-sulfur-coal applications have been noted to develop much heavier residual dustcake layers on the bags than those in low-sulfur-coal applications. It is not known whether similar tendencies will be experienced in spray dryer/fabric filter FGD systems installed on a high-sulfur-coal-fired source; however, it is important that such tendencies be evaluated in a pilot-scale facility that treats a flue gas from an actual high-sulfur-coal-fired source.

To investigate these types of concerns, EPRI has constructed and is operating a facility dedicated to FGD research for high-sulfur-coal utility applications. The facility, called the High Sulfur Test Center (HSTC), is located at the New York State Electric and Gas Company's Somerset station. Other sponsors of the facility include the Empire State Electric Energy Research Corporation, New York State Energy Research and Development Authority, Consolidation Coal Company, and the U.S. Department of Energy. The facility includes a 4-MW wet scrubber pilot unit, a 0.4-MW wet scrubber mini-pilot unit, and the subject of this paper, a 4-MW spray dryer FGD pilot unit. The facility began operation in April, 1987.

The spray dryer pilot unit there provides an opportunity to characterize spray dryer and particulate control device performance under actual high-sulfur-coal flue gas conditions. This will provide design information for utilities that are contemplating new or retrofit installations of this technology to power plants firing high-sulfur coal. Most of the testing with the spray dryer pilot unit has been with a reverse-gas fabric filter as the downstream particulate control device, but a limited amount of testing has been conducted with an ESP. More pilot-scale evaluation of spray dryer/ESP technology for high-sulfur-coal applications is being conducted in another EPRI co-funded program, by TVA at their Shawnee Test Facility (2).

The following sections of the paper provide a description of the spray dryer pilot unit process and equipment, then present results through July, 1988. The results presented document  $\text{SO}_2$  removal and particulate control performance for both the spray dryer/fabric filter and spray dryer/ESP configurations, although the majority of the results are for the spray dryer/fabric filter configuration. Other impacts of the spray dryer on each of the particulate control devices are also discussed.

## PILOT UNIT PROCESS DESCRIPTION

Figure 1 is a simplified process flow diagram for the spray dryer pilot unit. Flue gas is isokinetically extracted from the outlet duct on the Somerset station boiler. The coal fired there has a nominal 2.8 weight percent sulfur content and a 0.1 weight percent chloride content. This produces a flue gas  $\text{SO}_2$  concentration of about 1800 ppmv, and an HCl concentration of about 40 to 60 ppmv. The  $\text{SO}_2$  concentration of the spray dryer inlet gas is raised to 2000 ppmv by spiking with a small amount of  $\text{SO}_2$ . Further amounts of  $\text{SO}_2$  may be added to allow testing at pilot unit inlet  $\text{SO}_2$  levels of 2500 ppmv and greater. An electric heater is used to control the inlet flue gas temperature.

Flue gas enters the spray dryer, where it contacts a finely atomized slurry of fresh lime and recycled solids. The water content of the slurry evaporates, cooling the flue gas. Simultaneously, the lime in the slurry reacts with flue gas  $\text{SO}_2$ . Flue gas leaves the spray dryer from the side of the vessel, through an outlet duct which is centered in the bottom of the cone. The entrained solids containing equilibrium moisture levels continue to react with flue gas  $\text{SO}_2$ , and in fact  $\text{SO}_2$  removal in the particulate control device contributes significantly to the system  $\text{SO}_2$  removal. Gas leaving the particulate control device passes through an induced draft fan, then returns to the Somerset station duct.

As mentioned above, the feed to the atomizer is a mixture of lime slurry, recycle solids, and makeup water. The rate at which this combined slurry is fed to the atomizer is controlled to reduce the flue gas temperature at the spray dryer outlet to within a desired approach to adiabatic saturation. The fresh lime portion of this feed slurry is produced from pebble quicklime as a 24 weight percent solids content slurry, in a paste slaker. The fresh lime content of the feed slurry is controlled to result in the desired reagent ratio. The reagent ratio is defined as the moles of fresh lime in the atomizer feed slurry, neglecting any remaining lime in the recycled solids, divided by the number of moles of  $\text{SO}_2$  in the system inlet flue gas. Makeup water is added to the feed slurry because at a given reagent ratio the amount of water associated with the fresh lime slurry is generally not sufficient to reduce the flue gas temperature to the desired level. Solids collected in the particulate control device are recycled to raise the solids level in this combined feed slurry to 40 weight percent. This recycle is implemented both to improve overall lime utilization, and to improve droplet drying characteristics. Recycle of solids from the particulate control device is preferred, because solids which drop out in the spray dryer contain oversize chunks that tend to cause pluggage in the atomizer feed system.

## PILOT UNIT EQUIPMENT DESCRIPTION

The pilot unit is sized to treat 12,800 acfm of flue gas at 300°F. The spray dryer vessel has a 13-ft diameter and an 11-1/4-ft straight side, with a 45° cone bottom. The vessel volume of 2160 ft<sup>3</sup> results in a flue gas residence time of about 12 seconds when cooling 12,800 acfm of gas to within a 20°F approach to adiabatic saturation. The single rotary atomizer at the top of the vessel is equipped with a 250-mm (9.8-inch) diameter atomizer wheel with eight 6-mm (0.24 in.) diameter nozzles, and is driven by a variable-speed 50-hp motor. At the normal operating speed of 14,000 rpm, the tip speed at the outer edge of the wheel is about 600 ft/sec, which is within the range of commercial practice for utility spray dryer FGD systems.

The fabric filter particulate control device has five compartments, each with up to 36 bags in a 6 x 6 array. The woven fiberglass bags are 8 inches in

diameter and 22 ft long, are constructed of a 13.5 oz/yd<sup>2</sup>, 3 x 1 twill fabric, and have an acid-resistant finish. Although there are provisions for shake-deflate cleaning, all testing to date has employed reverse-gas cleaning. The time between compartment cleanings is set at 90 minutes. The cleaning consists of an initial 45-second null period, 30 seconds of reverse-gas flow at a rate of 1.5 acfm/ft<sup>2</sup>, and a final 30-second null period. With all 36 bags in each compartment, and a flue gas flow rate which results in the design 12-second residence time in the spray dryer, the air-to-cloth ratio at the fabric filter is about 1.4 acfm/ft<sup>2</sup>. During most of the pilot unit operation, the number of bags in each compartment has been reduced to raise the air-to-cloth ratio to 1.6 acfm/ft<sup>2</sup>. This higher value is thought to be more typical of utility practice for reverse-gas fabric filters.

The ESP particulate collector has five electrical fields available. Each field has 5 gas passages, and collecting plates that are 12 ft high and 12 ft long, with 12 in. plate spacing. The corona wires are 3/16-in. in diameter, with a rigid frame configuration, and the power supplies are rated at 65 kV and 200 mA. The rappers for both the plates and wires are electromagnetic. When the spray dryer is operating at the design conditions of 12,800 acfm at 300°F at the inlet, and spraying down to a 20°F at the outlet, the specific collection area (SCA) with all 5 fields in service is about 650 ft<sup>2</sup>/kacfm, and the flue gas velocity is about 3.3 ft/sec. The spray dryer tests have been conducted with 4 or less fields in service.

The pilot unit is well instrumented, with provisions to measure flue gas flow rate, SO<sub>2</sub> and O<sub>2</sub> concentrations at the spray dryer inlet, outlet, and particulate control device outlet, slurry flow rates, spray dryer and particulate control device pressure drop, and flue gas temperatures throughout the system. The fabric filter has had instrumentation added to allow tubesheet pressure drop and flue gas flow rates through each compartment to be measured, and to allow compartment wall temperatures to be monitored. Manual measurements are made on an hourly basis to measure flue gas wet bulb temperature, and on a less frequent basis manual measurements are made to measure flue gas particulate mass loadings and particle size distributions.

## RESULTS

A number of results from over a year of pilot unit operation are presented, for both the spray dryer/fabric filter and spray dryer/ESP configuration. First SO<sub>2</sub> removal, then particulate control performance results are presented. Although results are presented for both configurations, most of the data are for the spray dryer/fabric filter configuration. Also, a discussion is provided on the impacts of the spray dryer on the operation of the particulate control devices.

### SO<sub>2</sub> REMOVAL

#### Spray Dryer/Fabric Filter Configuration

Figure 2 summarizes typical SO<sub>2</sub> removal performance for the spray dryer/fabric filter system, with the data plotted as percent SO<sub>2</sub> removal versus reagent ratio. These data are for Mississippi Lime reagent and reflect operation of the spray dryer with an inlet SO<sub>2</sub> level of 2500 ppmv, an inlet temperature of 300°F, and with a 20°F approach to adiabatic saturation at the outlet. This inlet SO<sub>2</sub> level corresponds to a coal sulfur level of 3.5 to 4%. Also, these data correspond to operation with the normal Somerset station coal chloride level. The 0.1 weight percent coal chloride level produces HCl in the flue gas, which is absorbed



in the spray dryer and results in approximately a 0.4 weight percent chloride level in the spray dried solids.

Three curves are illustrated in Figure 2. The top curve represents overall system  $\text{SO}_2$  removal performance, and the curve just below represents the percent removal across the spray dryer. The bottom curve represents the fabric filter contribution to overall  $\text{SO}_2$  removal. This value is calculated by dividing the decrease in flue gas  $\text{SO}_2$  content across the fabric filter by the spray dryer inlet  $\text{SO}_2$  content. By defining this contribution in this manner, the spray dryer removal and the fabric filter contribution may be summed to get the overall system  $\text{SO}_2$  removal.

The results in Figure 2 illustrate  $\text{SO}_2$  removal performance for reagent ratio values from 1.0 to 1.6. The system  $\text{SO}_2$  removal varies from about 70% at a 1.0 reagent ratio to 90% at a 1.6 reagent ratio. The contribution to overall removal by the fabric filter appears to be important in achieving 90% removal. The  $\text{SO}_2$  removal across the spray dryer is limited to below 80%, while the fabric filter contribution amounts to about 11 to 16 percentage points.

The dashed line in Figure 2 represents 100% lime utilization. That is, it represents the  $\text{SO}_2$  removal that would result if all of the fresh lime added reacted to remove flue gas  $\text{SO}_2$ . At a reagent ratio of 1.0, 100% lime utilization would therefore correspond to 100% overall  $\text{SO}_2$  removal. The lime utilization values actually measured vary from about 70% at a 1.0 reagent ratio to 56% at a 1.6 reagent ratio.

At these high-sulfur-coal conditions, the source of quicklime slaked and used as fresh reagent appears to have measurable effect on system  $\text{SO}_2$  removal performance. This effect is illustrated in Figure 3, which summarizes overall system  $\text{SO}_2$  removal performance for 5 different sources of high calcium, high reactivity (as defined by ASTM 3-minute temperature rise criteria) quicklimes. The data plotted in the figure illustrate that with changes in lime source the reagent ratio required to achieve 90% overall  $\text{SO}_2$  removal varies from about 1.3 to 1.6. In fact, even variations in reactivity from delivery to delivery of the Mississippi Lime reagent may contribute to the data scatter seen in Figure 2. A number of possible explanations have been explored as to why similar quicklimes have such a wide variability in effectiveness as a high-sulfur spray dryer FGD reagent. An extensive amount of chemical and physical characterizations have been conducted on the limestones that are calcined by the lime plants, the quicklimes received at the HSTC, and the slaked lime that is fed to the pilot unit. However, no consistent explanation has been discovered to date.

As well as the choice of lime used, the spray dryer inlet conditions can markedly impact the  $\text{SO}_2$  removal performance of the system. In Figure 4, the  $\text{SO}_2$  removal performance for the spray dryer/fabric filter system is plotted for the most reactive of the 5 quicklimes from Figure 3, and for a lower inlet  $\text{SO}_2$  level of 2000 ppmv. A 2000 ppmv inlet  $\text{SO}_2$  level corresponds to a coal with approximately a 3% sulfur content. With this lime, from the Bellefonte Lime Company, and the lower 2000 ppmv inlet  $\text{SO}_2$  level, 90% overall  $\text{SO}_2$  removal is achieved at a reagent ratio of approximately 1.0. The resulting lime utilization is between 90 and 100%. This level of reagent utilization is similar to that of wet lime scrubbers applied to high-sulfur-coal sources.

The flue gas temperature at the spray dryer inlet can also markedly impact the system overall  $\text{SO}_2$  removal performance. Figure 5 summarizes the results of tests conducted with slaked Marblehead Lime, from their Pleasant Gap plant (now owned by Centre Lime), for operation at a 2500 ppmv inlet  $\text{SO}_2$  level. With a 300°F spray dryer inlet temperature, the overall  $\text{SO}_2$  removal just reaches 90% at a 1.6



reagent ratio. When the spray dryer inlet temperature is raised to a relatively high value of 350°F, the overall SO<sub>2</sub> removal increases dramatically to 98% at a 1.6 reagent ratio. An overall removal level of 90% is achieved at about a 1.2 reagent ratio with this lime at these inlet conditions. Thus, a 50°F increase in the spray dryer inlet temperature resulted in a 25% decrease in the quantity of lime required to achieve 90% overall SO<sub>2</sub> removal.

#### Spray Dryer/ESP Configuration

The ability of a spray dryer FGD system to achieve overall SO<sub>2</sub> removal levels of 90% and greater is dependent on the contribution to overall removal made by the particulate control device. The magnitude of this contribution is impacted by the type of control device used. This is illustrated in Figure 6, which compares the results of two tests conducted in the spray dryer/ESP mode to results from operation in the spray dryer/fabric filter mode. All of these tests were conducted with a 300°F spray dryer inlet temperature, a 2500 ppmv spray dryer inlet SO<sub>2</sub> level, a 20°F approach temperature, and with slaked Mississippi Lime reagent. The spray dryer/fabric filter results were previously illustrated in Figure 2.

The results in Figure 6 show that the overall SO<sub>2</sub> removal for the two tests with the ESP particulate collector is about 3 to 6 percentage points lower than with the fabric filter collector. As expected, the percent removal in the spray dryer is virtually identical in the two cases; the difference is in the contribution by the particulate control device. The contribution by the ESP ranges from 10 to 12 percentage points, while that for the fabric filter ranges from about 12 to 16 percentage points at these same conditions. While this difference in SO<sub>2</sub> removal by the particulate control device appears to be relatively minor, at these conditions it makes the difference in whether or not 90% overall SO<sub>2</sub> removal is achieved at a reagent ratio of 1.6. More SO<sub>2</sub> removal data for a spray dryer/ESP FGD system in a high-sulfur-coal application are presented in a previous paper (2), which summarizes results from another EPRI co-funded pilot-scale research project.

#### PARTICULATE CONTROL PERFORMANCE

The flue gas entering the spray dryer contains a mass loading of 2 to 4 gr/dscf. The particulate loading at the outlet of the spray dryer (or particulate control device inlet) has been measured to vary from 7 to over 15 gr/dscf, depending on spray dryer operating conditions. Because of the lime and recycle solids added at the spray dryer, the mass loading of the flue gas entering the particulate control device is about 4 to 8 times that of a typical flue gas from a pulverized-coal-fired boiler. The spray dryer also cools the flue gas, increases the moisture content and relative humidity, and correspondingly lowers the volumetric flow rate. The following discusses the impacts of these changes on the particulate control performance of the fabric filter and ESP.

#### Fabric Filter Performance

The reverse-gas fabric filter has been found to be a very effective particulate control device, in spite of the changes in flue gas conditions made by the spray dryer. Fabric filter outlet mass concentrations have been measured to vary from 0.0002 to 0.0013 gr/dscf during spray dryer operation. These concentrations correspond to removal efficiencies of 99.98% to over 99.99%, and the emission rates that are at least an order of magnitude below current NSPS requirements of 0.03 lb/10<sup>6</sup> Btu. Opacity measurements at the fabric filter outlet indicate values below 2%, that likely represent the accuracy of the zero calibration of the monitor rather than any measurable flue gas opacity. Cleaning spikes, when a just-cleaned compartment is put back in service, raise the opacity readings above

baseline by about 1% for several seconds. Also, when fabric filter upsets such as torn bags or a bag loose from a thimble occur, the outlet opacity can increase to 10% or considerably higher.

### ESP Performance

The particulate control performance of the ESP was measured under baseline conditions, with normal boiler fly ash entrained in 300°F flue gas, and then with the spray dryer in operation. Although the ESP has five electrical sections or fields, baseline ESP operation for these tests used only four fields. Using four fields lowered the SCA values to nearly match those in another, EPRI co-funded pilot evaluation of high-sulfur-coal spray dryer/ESP operation, at the TVA Shawnee test facility. Results from that test program were presented earlier in this session (2).

Particulate control results for the ESP at the High Sulfur Test Center are summarized in Table 1. Under baseline, 300°F operation, the SCA with four fields in operation averaged 348 ft<sup>2</sup>/kacfm, and the velocity through the ESP averaged 4.6 ft/sec. The inlet mass loading averaged 1.19 gr/acf, while the outlet averaged 0.0009 gr/acf (0.0013 gr/dscf). This corresponds to a particulate collection efficiency of 99.92%, and an outlet emission rate an order of magnitude below the current 0.03 lb/10<sup>6</sup> Btu NSPS limit.

With the spray dryer in operation, the net effect of the cooling of the flue gas from 300°F to 145°F, and the moisture added, lowers the volumetric flue gas rate sufficiently to raise the SCA to 409 ft<sup>2</sup>/kacf, and to lower the velocity to 3.9 ft/sec. These changes alone should improve ESP control effectiveness. The cooling increases the gas density in the ESP, which makes higher electrical field strengths possible, and decreases the gas viscosity, which would make the particles migrate to the plates more readily. Furthermore, particle size distribution measurements indicate that the fly ash entering the spray dryer pilot unit has a mass mean physical diameter of just less than 10 microns, whereas the particulate matter leaving the spray dryer when it is in operation has a mass mean physical diameter between 15 and 20 microns. The larger particles added in the spray dryer should be easier to collect than the smaller fly ash particles. All of these effects should result in improved particulate removal efficiency across the ESP when the spray dryer is in operation, although not necessarily lower emission rates.

The results in Table 1 show that the particulate removal efficiency in fact remains constant when the spray dryer is placed in service. The spray dryer raises the mass loading at the ESP inlet by a factor of 5 relative to the fly-ash-only value. Correspondingly, the outlet emissions increase by a factor of 5 when the spray dryer is in service, to 0.0045 gr/acf (0.0062 gr/dscf). The outlet emission rate is increased to 0.012 lb/10<sup>6</sup> Btu, which is still lower than the current NSPS limit. This relationship of seeing essentially no change in ESP percent particulate removal efficiency when the spray dryer is placed in service is similar to that seen at the TVA 10-MW spray dryer/ESP pilot unit at Shawnee (3). The reason the expected improvement in efficiency is not realized has been attributed to non-ideal scouring effects. Laboratory measurements indicate that the resistivity of the spray dried solids at the low temperatures and high flue gas relative humidity conditions in the ESP are in the range of 10<sup>8</sup> ohm-cm. Apparently this resistivity is low enough that electrostatic forces are not sufficient to avoid re-entrainment of collected particles by flue gas flowing past the plates.

ESP performance was observed to degrade significantly as the number of operating fields and hence the SCA was lowered. With 3 fields in operation, and a corresponding SCA of 312 ft<sup>2</sup>/kacfm, the outlet emissions during spray dryer

operation increased to about 0.015 gr/acf (0.021 gr/dscf), for a control efficiency of 99.76%. This outlet loading corresponds to 0.038 lb/10<sup>6</sup> Btu, which exceeds the current NSPS. With two fields, and an SCA of 211 ft<sup>2</sup>/kacfm, the outlet loadings averaged 0.074 gr/acf (0.101 gr/dscf), and the control efficiency was lowered to 99.16%. This outlet loading corresponds to an emission rate of 0.18 lb/10<sup>6</sup> Btu, which exceeds even the earlier, 1971 NSPS limit of 0.1 lb/10<sup>6</sup> Btu. These results suggest that spray dryer retrofits to units with small existing ESPs (e.g., less than 300 SCA) may have difficulty maintaining emissions control compliance.

Baseline opacity for the flue gas at the ESP outlet remained below 10%, even with only 2 fields operating. With 4 fields operating, rapping spikes had a negligible effect on outlet opacity. With only 2 fields operating, spikes of up to 70% opacity were encountered during plate raps. These spikes were reduced to below 20% by lowering the plate rapping intensity from 10 ft-lb to only 2 ft-lb.

It should be noted that since the performance of the ESP appears to be limited by a non-ideal scouring effect, and the amount of scouring may be influenced by the flue gas velocity through the ESP, the emissions control performance data presented here may be specific to this flue gas velocity. Thus, these results may overpredict the performance of ESPs that operate at higher flue gas velocities, and underpredict the performance of ESPs that operate at lower velocities. Furthermore, this pilot ESP has plates that are not as tall as most full-scale units, but similar in length. This gives the pilot ESP at the HSTC a relatively high aspect ratio (effective plate length divided by effective plate height). Higher aspect ratios are known to reduce re-entrainment emissions, so for this reason these pilot-scale data may overpredict the performance of full-scale ESPs with similar SCA values. This is thought to be one reason why the removal efficiencies in Table 1 are somewhat higher than those measured at similar conditions in the TVA pilot unit. Those results were discussed earlier in this session (2). The ESP at the TVA pilot unit has plates that are almost twice as tall as those at the HSTC but about the same length, and hence, have a much lower aspect ratio.

#### PILOT UNIT OPERATING EXPERIENCE

In addition to determining SO<sub>2</sub> removal and particulate removal performance for a spray dryer/fabric filter or spray dryer/ESP in high-sulfur-coal operation, a further objective of this pilot testing is to determine how well the spray dryer FGD system operates under these conditions, and what effects the spray dryer has on the operation of the particulate control device. The operation of the spray dryer FGD system, fabric filter, and ESP are discussed separately below.

#### SPRAY DRYER FGD SYSTEM OPERATION

In general, the spray dryer FGD system has operated well. The following will discuss the atomizer feed preparation system, the spray dryer itself, and the solids handling system.

##### Atomizer Feed Slurry Preparation System

Operation under high-sulfur-coal conditions raises the lime content of the atomizer feed to the point that it may represent as much as 45% of the solids present. In spite of the relatively high proportions of the viscous, 24 weight percent solids content lime slurry in the atomizer feed, few problems have arisen in mixing a 40 weight percent solids content atomizer feed slurry. The main problems have been with buildups of solids in the bottom of the mix tanks, pluggage of



pump suction lines, and pluggage of the atomizer slurry distributor. Most of these problems had rather site-specific resolutions, such as increased mix tank agitator impeller size and motor horsepower, going to fewer, larger openings in the slurry distributor, etc. However, it was noted that the incidences of feed line pluggage were reduced when the solids recycle scheme was modified to recycle only particulate control device solids rather than a mixture of spray dryer and particulate control device solids. The spray dryer solids have more large particles that tend to plug the slurry strainers.

### Spray Dryer Operation

The spray dryer vessel flue gas residence time of 10 to 12 seconds has proven adequate to dry the 40 weight percent solids content feed slurry. There have been no incidences of excessive buildup of damp solids on the vessel roof or side walls. There have been a few instances where the cone bottom has plugged with solids, but these somewhat isolated incidences do not appear to be related to inadequate solids drying. Instead, those pluggages appear to result when larger pieces of the normal wall deposits occasionally break loose. It should be noted that the bottom of this vessel does not have any sort of lump breaker installed, as do many full-scale spray dryer FGD system vessels.

The rotary atomizer drive unit has been relatively trouble free as well, in spite of the numerous start-ups and shutdowns associated with pilot unit operation. The atomizer wheels have had a tendency for erosion, both on the floor of the wheel which is in direct slurry contact, and on the underside of the wheel that is exposed only to flue gas. It should be noted that this pilot-scale wheel is constructed of a relatively soft 329 stainless steel. Unlike atomizer wheels for most full-scale installations, this wheel does not have any sort of ceramic wear plates, other than the silicon carbide nozzle inserts. The wheel manufacturer has applied a proprietary coating on the floor of the wheel, but this has proven relatively ineffective in controlling erosion by the abrasive recycle/lime slurry. No wheel has failed due to this erosion wear, but none has seen more than a few thousand hours of operation.

### Solids Handling System

The solids collected in the bottom of the spray dryer are moved by an inclined screw conveyor to a surge vessel for transfer to a vacuum pneumatic transport system. Other than the occasional pluggage of the spray dryer bottom as described above, the only problem with this system has been site-specific screw conveyor failures.

The solids collected in the fabric filter are evacuated from each compartment hopper by a vacuum pneumatic conveying system. The compartments are isolated from the vacuum system by air-actuated knife gate valves. These valves frequently require maintenance, because of a buildup of solids on the track and seat for the knife blade. The buildups cause the knife blades to stick, generally in the closed position, and/or disturb the seating of the valves so that they do not seal properly. These problems are resolved by removing and cleaning the valve internals. It is apparently the relatively cohesive nature of the spray-dried solids which causes this problem. The solids contain 0.5 to 1.0 weight percent moisture, and almost always contain excess lime. Both of these constituents can cause cementation reactions that make the solids more cohesive than normal fly ash.

Full-scale spray dryer FGD systems, all in low-sulfur-coal applications, employ a variety of solids handling system designs. Vacuum pneumatic, mechanical, and pressure pneumatic (dilute- and dense-phase transport) systems are all used in the full-scale systems. Most utility spray dryer FGD systems report solids



handling system problems (4), almost independent of the type of handling system used. Based on the experiences at this pilot unit, solids handling will likely be troublesome in high-sulfur-coal applications of this technology as well. A future EPRI research effort is planned to develop an optimum solids handling system design that would minimize such problems for these FGD systems.

### FABRIC FILTER OPERATION

There were several anticipated impacts of the spray dryer FGD system on the operation of the fabric filter, such as on pressure drop performance, filter bag residual dustcake weights, bag material life, and on tendencies for metal corrosion within the fabric filter. Each of these is discussed below.

#### Fabric Filter Pressure Drop

The fabric filter has been in service about 8000 hours downstream of the operating spray dryer, and has undergone approximately 60 start-up and shutdown cycles. Throughout this period, spray dryer operating conditions have changed frequently as short term (3-day) SO<sub>2</sub> removal tests have been conducted. During facility start-up, approximately the first 1700 hours of operation, the fabric filter was operated with all 36 bags in each compartment, resulting in an air-to-cloth ratio of about 1.4 acfm/ft<sup>2</sup>. The cleaning program was set for time or pressure drop initiation, such that the time between compartment cleaning varied between 1 and 3 hours. After this start-up period, the fabric filter operation was changed to more closely reflect typical conditions for fabric filters in utility high-sulfur-coal service. It was changed to operate at an air-to-cloth ratio of 1.6 acfm/ft<sup>2</sup>, and with 90 minutes between compartment cleanings. To minimize variations in flange-to-flange pressure drop with time, a compartment cleans every 18 minutes, such that all 5 are cleaned every 90 minutes.

Figure 7 summarizes flange-to-flange pressure drop performance for the fabric filter for over 6000 hours of operation downstream of the spray dryer. These data are for the period after the fabric filter air-to-cloth ratio was raised to 1.6 acfm/ft<sup>2</sup>, and the time between compartment cleaning was set at 90 minutes. Most of the pressure drop data in the latter period of the graph (April through June, 1988) lie within the range of 5 to 6 in. H<sub>2</sub>O. A few data points exceed 7 in. H<sub>2</sub>O, but these are generally just after the fabric filter has come on line after an outage. In each of these instances, the flange-to-flange pressure drop quickly recovers. The entrance and exit pressure losses average about 1.5 in. H<sub>2</sub>O for this fabric filter. Thus, tubesheet pressure drop values for the latter time period would be about 3.5 to 4.5 in. H<sub>2</sub>O. Both the flange-to-flange and tubesheet pressure drop data for this pilot fabric filter are within the range of typical pressure drop performance for conventional utility reverse gas fabric filters (5). This indicates that there has been no significant adverse effect of the spray dryer on fabric filter pressure drop performance. Remember that these data represent pressure drop performance after a total of only 8000 hours of operation on the bags in the two periods described above. Also, these pressure drop data reflect numerous start-ups and shutdowns as well as frequent changes in spray dryer conditions while short term SO<sub>2</sub> removal tests are being conducted.

#### Bag Residual Dustcake Weights

Bag weights were measured in the five compartments after 10, 12, and 14 months of fabric filter operation. Each time, six of the bags in each compartment were weighed. The same bags were weighed each time, and the six bags were in equivalent positions within each compartment. The weight gains per bag (above the weight of a new bag) averaged less than 20 lb. Equivalent residual dustcake areal loadings (calculated as the weight gain per bag divided by the bag filtering area)

averaged just under 0.4 lb/ft<sup>2</sup>. These average areal loading values are very similar to values measured after long-term operation of the fabric filter downstream of the pilot spray dryer at the low-sulfur-coal-fired Arapahoe facility, and are only about one-half of steady-state values measured for a reverse-gas fabric filters in conventional high-sulfur-coal service (6). These data suggest that fabric filters downstream of spray dryers in high-sulfur-coal service will not experience high residual dustcake weights. Of course, these data represent just over one year of operation under these conditions, and may not represent the residual dustcake weights that would be experienced after several years of operation.

#### Bag Fabric Life

Bag samples were cut from single bags in the fabric filter on two occasions, after 9 and 14 months of fabric filter operation. In each instance the bag which was cut was replaced. Fabric sections from the top, middle, and bottom of each bag sample were subjected to Mullen Burst Strength measurements. Also, samples of new bag material were subjected to these measurements at the same time. The burst pressures for the aged bags averaged 64% of a new bag value for the samples reflecting 9 months of operation, and 45% for the sample reflecting 14 months. The bag tester stated that these were normal strength retention values for bags in service 9 and 14 months, respectively. This is taken as an indication that the 13.5 oz. fiberglass fabric and acid-resistant finish are not adversely affected by this high-sulfur-coal spray dryer application.

#### Wall Corrosion

There were, however, negative impacts of the spray dryer on the fabric filter. One was a tendency for corrosion on the mild steel compartment walls. In addition to increasing the mass loading in the flue gas going to the fabric filter, the spray dryer cools and humidifies the gas. For a spray dryer application, the flue gas will be as little as 20°F above its moisture dew point, while in conventional fabric filter applications the flue gas is about 200°F above. With the flue gas so close to its moisture dew point, any significant cooling in the fabric filter will result in condensation on compartment walls. Typical causes of cooling are ambient air leaks around compartment entry doors, inadequate thicknesses or incomplete coverage of insulation, and penetrations of metal braces, etc., through the insulation to the compartment walls. As any condensation occurs, equilibrium concentrations of SO<sub>2</sub> will absorb into the moisture. Once absorbed, this sulfurous acid will be partially oxidized to sulfuric acid. Because sulfuric acid forms an azeotrope with water, the condensed moisture will not evaporate once the wall temperatures are raised above the flue gas moisture dew point. Furthermore, since the compartment walls are on the "clean" side of the bags, there is no lime present to neutralize the acid formed. This acid would be expected to accelerate corrosion of the mild steel compartment walls.

Significant corrosion was noted in the fabric filter compartment downstream of the pilot spray dryer at Arapahoe (7). That fabric filter was designed to treat normal 300°F flue gas, and so had only 2 inches of wall insulation. Thermocouples installed on compartment walls confirmed that during periods of low ambient temperatures, the wall temperatures were dropping below the flue gas moisture dew point. The compartment walls were sandblasted and a protective coating was applied. Although it was anticipated that corrosion would be experienced in the fabric filter downstream of the spray dryer at the HSTC, this unit is more appropriately insulated with a 6-inch thickness of fiberglass batts. It was decided to start with uncoated walls at the HSTC to allow evaluation of the extent of the corrosive tendencies.

However, corrosion has been experienced on the compartment walls of the fabric filter at the HSTC. The corrosion appears as large areas of exfoliating oxidation products, which tend to flake and fall from the walls. As at Arapahoe, during periods of low ambient temperatures the compartment wall temperatures have been measured at several degrees below the flue gas moisture dew point. Corrosion is worst near the lower compartment doors, where door seal leaks are obviously occurring. The corrosion also appears worst in end compartments, which have three rather than two walls that are exposed to ambient temperatures. Wall thickness measurements made after nearly one year of operation indicated that metal losses on compartment doors amount to as much as 10% of the original thickness. On compartment walls, metal losses were limited to a few thousandths of an inch.

Of course, a pilot-scale fabric filter represents a worst case for such corrosion, because of the relatively high surface area to volume ratio of the small compartments. However, similar corrosion tendencies have been noted in full-scale fabric filters in low-sulfur spray dryer applications (7). One full-scale spray dryer/fabric filter installation has had protective coatings applied to practically all of the fabric filter compartment wall surfaces (8).

Several actions have been taken to slow or eliminate this corrosion. First, shaker mechanisms (for later shake-deflate cleaning tests) which penetrated the wall of each compartment were removed, and the exposed areas where they were mounted were insulated. This greatly improved the integrity of the insulation coverage. Also, improved door seal materials are being evaluated in an effort to reduce or eliminate ambient air inleakage. Finally, the entire fabric filter will be sandblasted and a number of protective coatings will be applied to evaluate their relative effectiveness.

#### Bag Clamp Failures

In addition to wall corrosion, numerous fabric filter bag clamp failures have been experienced. When the bags were originally installed in March, 1987, they were attached to the tubesheet thimbles with stainless steel screw clamps. Within a matter of months, these clamps began to fail, allowing the bags to come loose from their tubesheet thimbles. At first it was thought that the clamps were failing because of overtightening during installation. However, replacement clamps that were properly tightened began to fail after only a few months in service as well.

The failures have been attributed to chloride-induced stress corrosion cracking. Similar failures were experienced in pilot testing at the EPRI Arapahoe test facility (9). There, stainless steel bag clamps were observed to fail in the fabric filter downstream of the pilot spray dryer during and after tests where calcium chloride was added to the atomizer feed slurry as an SO<sub>2</sub> removal performance additive. A failure analysis on those clamps confirmed chloride-induced stress corrosion cracking as the mode of failure. Although the clamps are on the "clean" side of the bags, apparently enough of the solids bleed through the bags to cause chloride deposition on the clamps in the moist flue gas environment there. At the HSTC, though, no such performance additive testing was conducted. Instead, the chlorides appear to be coming from the flue gas. The coal fired at the Somerset station has approximately a 0.1 weight percent chloride content. This produces about 40 to 60 ppmv HCl levels in the flue gas. This in turn is absorbed in the spray dryer FGD system, producing similar calcium chloride levels to those in the performance additive tests at Arapahoe.

It is apparent that bags clamps made of 300-series stainless steel will not be suitable for use in a fabric filter downstream of a spray dryer treating flue gas with an appreciable chloride content. Two alternative clamps have been



evaluated at the HSTC. One novel approach has been to use Teflon cable ties to fasten the bags to the thimbles. While these cable ties were not subject to chloride attack, it was difficult to keep the ties tight enough to avoid having bags work their way loose from the thimbles. Another alternative was cadmium-plated stainless steel clamps. However, these clamps failed much quicker than even plain stainless steel clamps. Apparently the plating was partially scraped off during clamp installation, resulting in small electrolytic cells of dissimilar metals that promoted more rapid corrosion. High nickel content alloys are typically used in high chloride concentration environments, but no ready source of bag clamps of such alloys has been identified.

#### ESP OPERATION

The spray dryer/ESP configuration was operated for only one month, so little can be said about the impacts of the spray dryer on ESP operation. During this short period no corrosion tendencies, or other negative impacts of the spray dryer on the ESP were noted. In fact one improvement was noted, in flue gas electrical properties, during spray dryer operation. This improvement is largely due to the increased gas density which results from the cooling which occurs in the spray dryer. As an example of the improvement, the first-field electric field strength was increased by about 10% with the spray dryer in operation, relative to the field strengths measured at the same current density without the spray dryer in operation.

#### FUTURE TESTING

Currently, the spray dryer FGD pilot unit is shut down while duct injection pilot tests are being conducted. When spray dryer FGD pilot unit testing resumes in 1989, more short-term SO<sub>2</sub> removal tests will be conducted. These tests will continue to evaluate the interactions between lime source, inlet SO<sub>2</sub> level, inlet temperature, and reagent ratio. The effects of coal chloride level on SO<sub>2</sub> removal performance will be another major variable evaluated. Also, these short term tests will evaluate the effects of fabric filter type and air-to-cloth ratio on SO<sub>2</sub> removal performance (i.e., a reverse-gas fabric filter at a low air-to-cloth ratio versus a pulse-jet fabric filter at an air-to-cloth ratio near 4 acfm/ft<sup>2</sup>). These tests are intended to define the capabilities of the process over a wide range of operating conditions, to explore improvements that may reduce SO<sub>2</sub> control costs, and/or expand the applicability of the process to coals with higher sulfur levels.

After the short-term tests are completed, long-term (e.g., several month) tests will be conducted to both characterize and optimize fabric filter operation downstream of a spray dryer FGD system. For the reverse-gas fabric filter, variables are to include air-to-cloth ratio, bag fabrics, time between compartment cleanings, sonic-horn assistance, and others. For the pulse-jet fabric filter, variables evaluated likely include air-to-cloth ratio, cleaning frequency and duration, and others. Both of these series of tests will be aimed at establishing whether smaller, less expensive reverse-gas or pulse-jet fabric filters can be substituted for the rather conservatively sized (low air-to-cloth ratio) reverse-gas fabric filters that are now commonly used in spray dryer FGD systems.

Finally, more tests in the spray dryer/ESP configuration, and screening tests in a spray dryer/shake-deflate-cleaned fabric filter configuration will likely be conducted.



## SUMMARY

After over a year of operation of the EPRI HSTC spray dryer pilot unit under actual high-sulfur-coal operating conditions, the following observations may be made:

- With the baseline, Mississippi Lime reagent, spray dryer inlet conditions of 2500 ppmv SO<sub>2</sub> concentration and 300°F, and operation at a 20°F approach to adiabatic saturation, 90% overall SO<sub>2</sub> removal can be achieved with a spray dryer/fabric filter FGD system at a reagent ratio near 1.6. However, the reagent ratio required to achieve 90% overall SO<sub>2</sub> removal may be greatly reduced at lower inlet SO<sub>2</sub> levels and higher inlet flue gas temperatures. Also, some limes which have proven to be more effective spray dryer reagents may achieve 90% overall removal at lower reagent ratios. It should be noted that these results are for operation with a coal chloride level of 0.1 weight percent. Higher or lower chloride levels will likely decrease or increase, respectively, the reagent ratio requirements to achieve a given level of SO<sub>2</sub> removal.
- With an ESP rather than a fabric filter particulate collection device, overall SO<sub>2</sub> is reduced by 3 to 6 percentage points relative to that with the fabric filter. While this does not appear to be a large penalty, it makes the difference as to whether or not 90% overall SO<sub>2</sub> removal can be achieved at a 1.6 reagent ratio under baseline 2500 ppmv, 300°F inlet conditions.
- Particulate removal by the fabric filter downstream of the spray dryer remained excellent in approximately one year of operation. In spite of the high inlet mass loadings caused by the material added in the spray dryer, outlet emission rates remained an order of magnitude lower than the current NSPS level of 0.03 lb/10<sup>6</sup> Btu. Similarly, outlet opacity values remained near zero.
- Particulate removal by the ESP did not increase in efficiency when the spray dryer was placed in service, as was expected. Under what was considered baseline ESP conditions, with 4 fields in service, the particulate control efficiency remained at 99.92% when the spray dryer was placed in service. The ESP inlet mass loading increased by a factor of 5 when the spray dryer was placed in service, and the outlet loading increased by a similar factor. Outlet emissions remained below 0.03 lb/10<sup>6</sup> Btu with 4 fields in service and an SCA of about 400 ft<sup>2</sup>/kacfm, increased to between 0.03 and 0.1 lb/10<sup>6</sup> Btu with 3 fields and a 300 SCA, and increased to above 0.1 lb/10<sup>6</sup> Btu with only 2 fields and a 200 SCA. Under spray dryer conditions, the ESP performance appeared to be limited by re-entrainment. Although rapping re-entrainment likely contributed to the emissions, the main limitation appears to be scouring of collected particles from the plates. This is speculated to result because of the low (10<sup>8</sup> ohm-cm) resistivity of the spray dried solids under the high flue gas relative humidity conditions. This results in reducing the electrical forces that tend to hold the collected particles to the plates. The amount of scouring that occurs is likely dependent on the flue gas velocity through the ESP, so the collection efficiency data cited above are probably specific to the 3.9 ft/sec velocity through the ESP in these tests. Also, the relatively short plate height for this pilot ESP,

relative to that of a full-scale unit with a similar SCA and flue gas velocity, is favorable for minimizing re-entrainment emissions. For this reason the collection efficiency data cited above may somewhat overpredict the performance of a full-scale unit.

- Operation of the spray dryer FGD system under high-sulfur-coal conditions has generally been quite successful, with the few problems being similar to those of existing units on low-sulfur-coal applications. Particularly, it appears that solids handling will be a problem in high-sulfur-coal applications of spray dryer FGD technology, as it has in existing low-sulfur-coal applications.
- The addition of a spray dryer upstream of a fabric filter in high-sulfur-coal applications does not appear to adversely effect pressure drop performance, in spite of the increased mass loading which results. Neither does bag fabric life appear to be adversely affected. Bag residual dustcake weights seem to be more like those of reverse-gas fabric filters in low-sulfur-coal spray dryer installations than the higher values that have been experienced in reverse-gas fabric filters in conventional high-sulfur-coal applications.

There do appear to be some adverse effects of the spray dryer FGD system on fabric filter operation. First, there appears to be a pronounced tendency for corrosion of mild steel compartment walls. These corrosion tendencies will have to be addressed by improved fabric filter insulation practices, and perhaps spot applications of protective coatings. Also, for high-sulfur coals with a significant chloride content (e.g. 0.1 weight percent or greater) there will be problems with stress corrosion cracking of 300-series stainless steel components such as bag clamps. The best solution to this problem is to employ alternate, chloride-resistant materials of construction.

To summarize, the results to date are promising for the applicability of spray dryer FGD technology for high-sulfur-coal applications. Particularly for new installations where a fabric filter will likely be installed as the particulate control device, 90% overall SO<sub>2</sub> removal and greater can be achieved, with little penalty to particulate control device performance. For retrofit situations, where an existing ESP is to be used, slightly lower SO<sub>2</sub> removal performance can be expected. The greater technical issue for such applications, though, is whether particulate control compliance can be maintained when the spray dryer is retrofitted.

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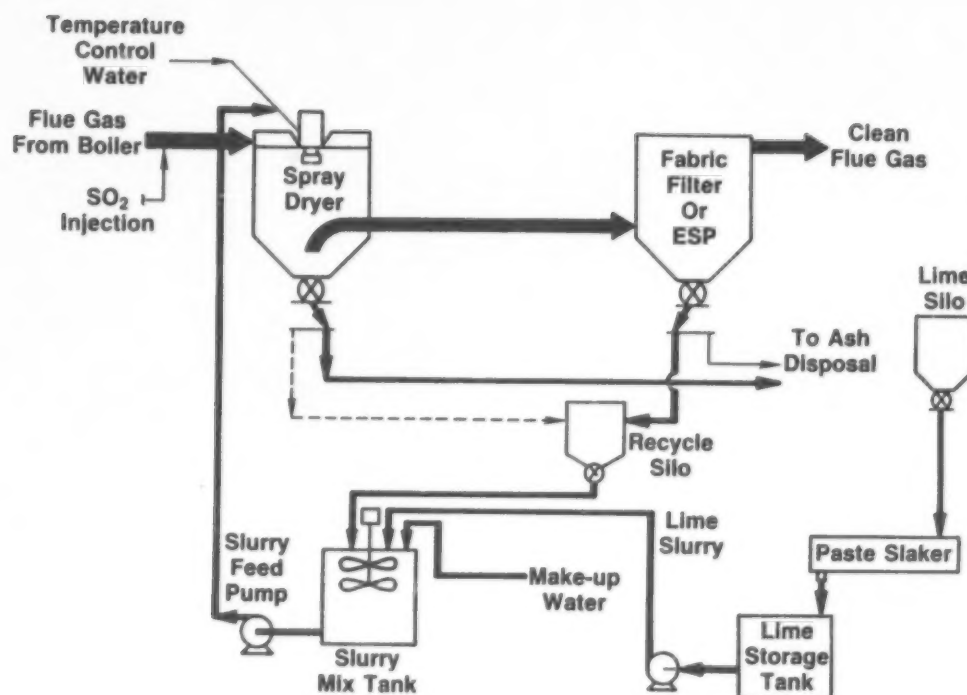


Figure 1. Process flow diagram for the HSTC spray dryer FGD pilot unit.

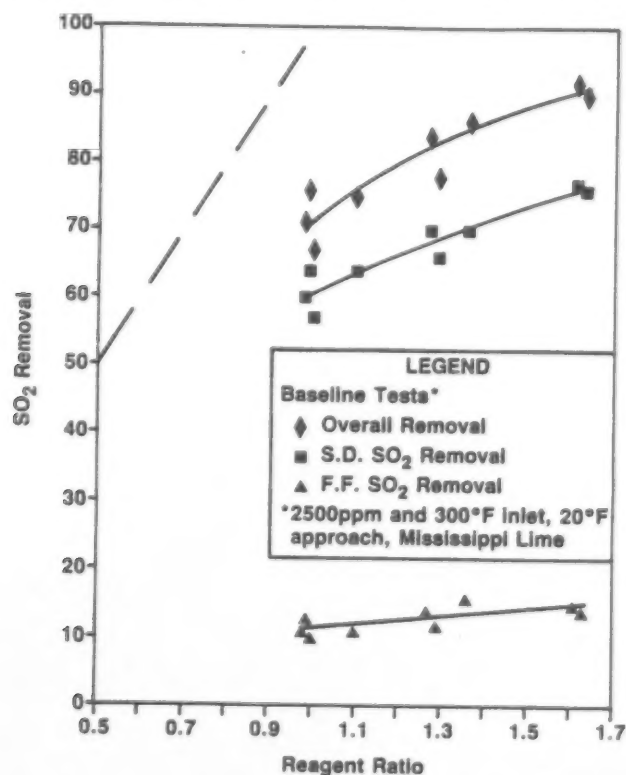


Figure 2. SO<sub>2</sub> removal versus reagent ratio for Mississippi Lime reagent, for a spray dryer inlet SO<sub>2</sub> level of 2500 ppmv, for operation with the fabric filter particulate collector.



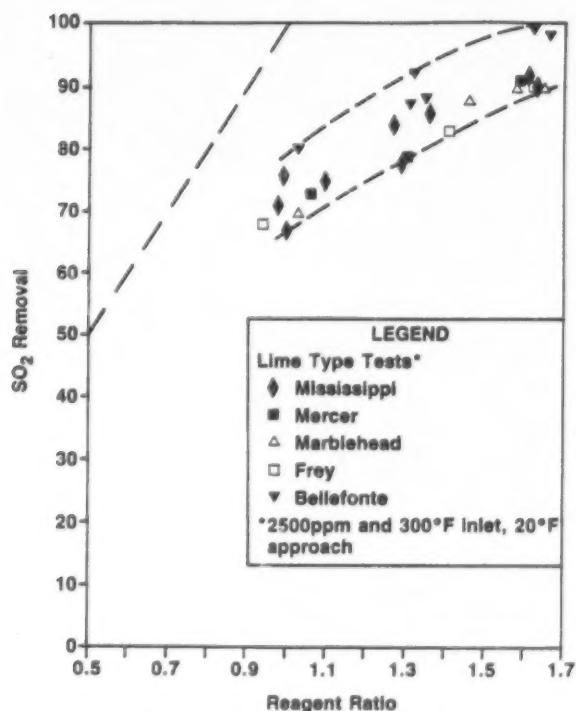


Figure 3. Overall SO<sub>2</sub> removal versus reagent ratio for five different limes, for a spray dryer inlet SO<sub>2</sub> level of 2500 ppmv, for operation with the fabric filter particulate collector.

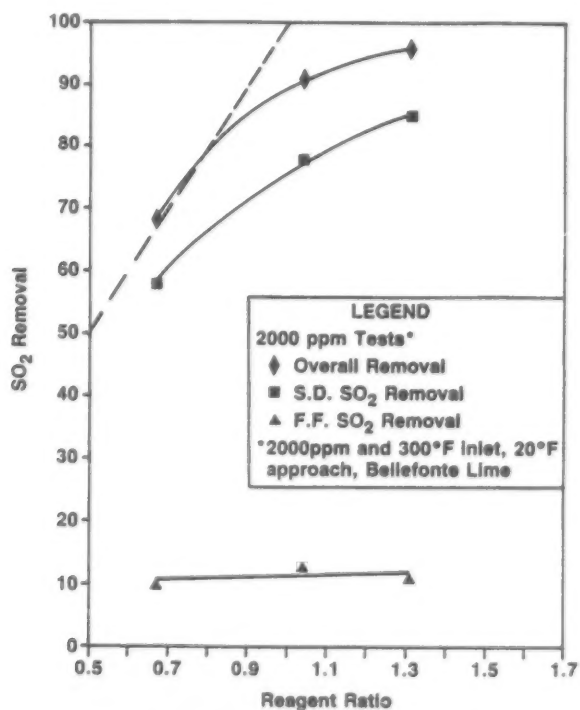


Figure 4. SO<sub>2</sub> removal versus reagent ratio for Bellefonte Lime reagent, for a spray dryer inlet SO<sub>2</sub> level of 2000 ppmv, for operation with the fabric filter particulate collector.

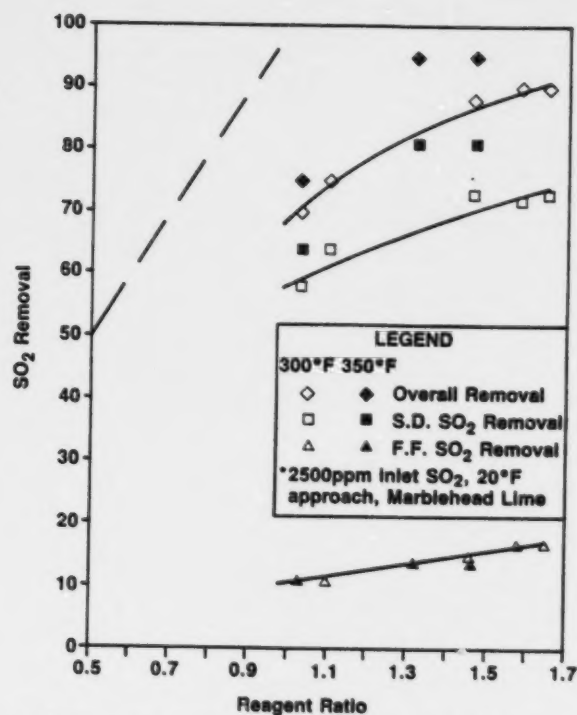


Figure 5. SO<sub>2</sub> removal versus reagent ratio for Marblehead Lime reagent, for a spray dryer inlet SO<sub>2</sub> level of 2500 ppmv, at two different inlet temperatures, for operation with the fabric filter particulate collector.

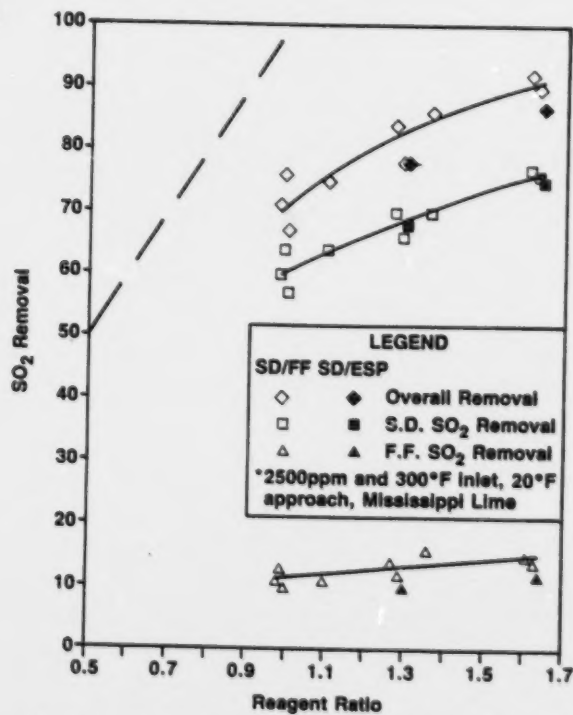


Figure 6. SO<sub>2</sub> removal versus reagent ratio for Mississippi Lime reagent, for a spray dryer inlet SO<sub>2</sub> level of 2500 ppmv, for operation with the ESP particulate collector, compared to that with the fabric filter particulate collector.

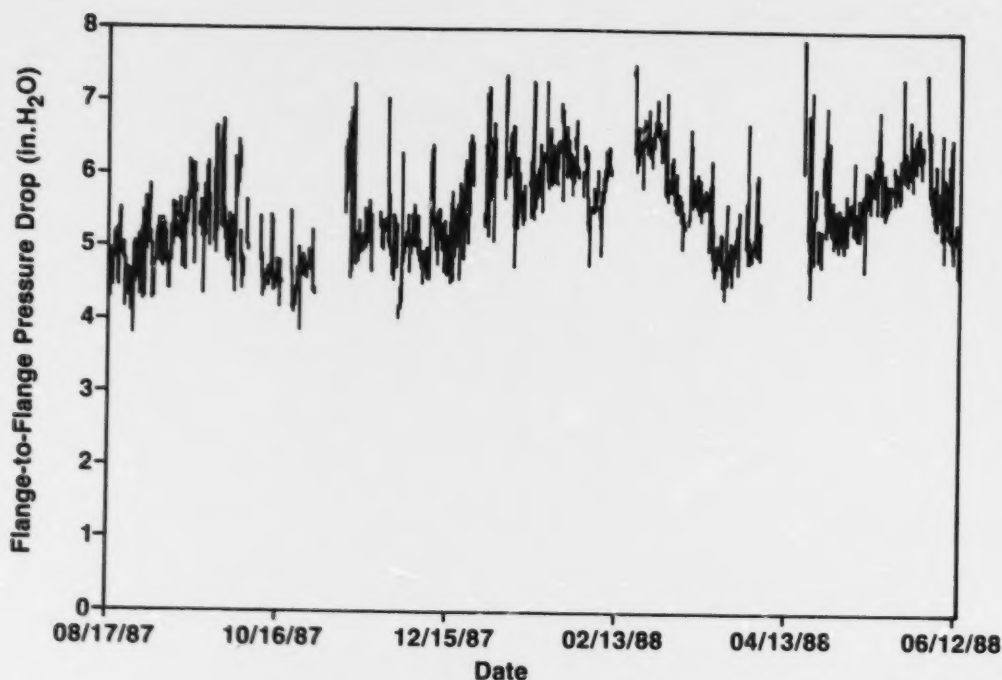


Figure 7. Flange-to-flange pressure drop data for the HSTC pilot fabric filter in operation downstream of the pilot spray dryer.

TABLE 1. SUMMARY OF ESP PARTICULATE CONTROL PERFORMANCE

| Parameter                                   | Without Spray Dryer<br>in Operation | With Spray Dryer<br>in Operation |
|---------------------------------------------|-------------------------------------|----------------------------------|
| Number of Fields in Operation               | 4                                   | 4                                |
| SCA (ft <sup>2</sup> /kacfm)                | 348                                 | 409                              |
| Flue Gas Velocity (ft/sec)                  | 4.6                                 | 3.9                              |
| Inlet Mass Loading (gr/acf)                 | 1.2                                 | 5.7                              |
| Outlet Mass Loading (gr/acf)                | 0.0009                              | 0.0045                           |
| (gr/dscf)                                   | 0.0013                              | 0.0062                           |
| Mass Emission Rate (lb/10 <sup>6</sup> Btu) | 0.0024                              | 0.012                            |
| Particulate Control Efficiency (%)          | 99.92                               | 99.92                            |
| Particulate Penetration (%)                 | 0.08                                | 0.08                             |

## SESSION 4A: FURNACE SORBENT INJECTION - DEMONSTRATIONS

Chairman, Richard D. Stern, EPA/AEERL

### OPERATION OF THE LIMB/HUMIDIFIER DEMONSTRATION AT EDGEWATER

by: Paul S. Nolan  
Babcock & Wilcox  
20 S. Van Buren Avenue  
Barberton, OH 44203

Robert V. Hendriks and Nicholas Kresovich  
U.S. Environmental Protection Agency  
Air and Energy Engineering Research Laboratory  
Research Triangle Park, NC 27711

### ABSTRACT

The LIMB Demonstration Project is being conducted as a full-scale demonstration of technology targeted for retrofit application for moderate control of SO<sub>2</sub> and NO<sub>x</sub> emissions from coal-fired utility boilers. Initial operation of LIMB at Ohio Edison's Edgewater Station in September 1987 showed that LIMB technology is capable of exceeding its SO<sub>2</sub> control goal of 50% removal and NO<sub>x</sub> emission goal of 0.5 lb/million Btu predicted by earlier research and development work.

The most significant operational problem encountered was associated with downstream particulate control. The high resistivity ash produced during sorbent injection significantly reduced Edgewater electrostatic precipitator (ESP) performance to the point that continuous operation of the injection system was limited to 3 to 8 hours per test.

A full-scale flue gas humidification system has been added to the Edgewater unit to supplement LIMB and came on-line in August 1988. The humidifier was designed to condition the flue gas to reduce ash resistivity as well as to increase overall SO<sub>2</sub> removal by promoting downstream sorbent/gas reaction.

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.



## INTRODUCTION

Control of emissions of sulfur dioxide and nitrogen oxides,  $\text{SO}_2$  and  $\text{NO}_x$ , from existing coal-fired utility boilers is a significant concern in the United States since these two pollutants are generally considered to be major precursors of acid rain. Coal-fired utility boilers account for about 65% of the  $\text{SO}_2$  and 29% of the  $\text{NO}_x$  emissions in this country (1).

One low-cost alternative for control of these emissions is LIMB (Limestone Injection Multistage Burner), which is based on the injection of a dry sorbent into the boiler for direct capture of  $\text{SO}_2$  in the flue gas, combined with the use of low- $\text{NO}_x$  burners for reduction of  $\text{NO}_x$  emissions. In 1981, the U.S. Environmental Protection Agency (EPA) initiated a research program to develop the LIMB process and bring the technology to a commercial scale. By 1985, the technology was sufficiently proven at bench and pilot scale to begin a full-scale demonstration of a LIMB system at the 104 MW Unit 4 boiler at Ohio Edison's Edgewater Station in Lorain, Ohio. EPA is sharing the cost of the project with the major participants: the State of Ohio, Ohio Edison, Babcock & Wilcox (B&W), and Consolidation Coal.

The demonstration project consists of several distinct phases: a preliminary phase to develop the LIMB process design specifically for the host boiler, a construction and start-up phase, and an evaluation phase. The process design was completed and the basic LIMB system was installed at Edgewater by mid-1987. The system was started up in July 1987 and was operated intermittently through September 1987, when Unit 4 was shut down for a scheduled maintenance outage. Meanwhile, research at EPA had led to development of a downstream flue gas humidification step which has the effect of increasing  $\text{SO}_2$  removal through the utilization of unreacted free lime in the LIMB ash. Because of this technological development, the LIMB demonstration contract was expanded to include the addition of a humidifier at Edgewater to supplement the basic LIMB sorbent injection system. As will be discussed later, the humidifier will also condition the flue gas to restore lost efficiency in the downstream ESP experienced during sorbent injection. The humidifier began operation in August 1988 and is currently an integral part of the LIMB demonstration being evaluated at Edgewater.

## LIMB DESIGN

In the LIMB process a calcium-based sorbent is injected into the boiler where it is calcined to active calcium oxide, and then reacts with sulfur dioxide and oxygen in the flue gas to produce calcium sulfate. The product is a solid which is removed with ash in the particulate collection device. Research has shown that maximum sorbent reactivity and sulfation is obtained in the temperature range of roughly 1600 to 2300°F, so the injectors are located where the temperature in the boiler is at the upper end of this range. The  $\text{NO}_x$  control component of LIMB is implemented by the use of low- $\text{NO}_x$  burners which keep flame temperatures relatively low to reduce  $\text{NO}_x$  formation. A generalized process diagram is shown in Figure 1. A brief description of the Edgewater equipment is provided here as an aid to the reader in understanding later operating results. A more detailed review of LIMB design issues and the basis for the Edgewater design was presented in an earlier paper (2).

At Edgewater, commercial calcitic hydrated lime is delivered to the plant by truck, stored in a silo, and conveyed to the boiler through a differential weight loss feeder which provides an accurate feed rate. A flexible sorbent injection scheme has been installed consisting of three rows of injectors on the front wall at and just above the nose of the boiler (these rows are designated by their elevation--181, 187, and 191 ft). Two sets of feeders, conveying lines, and distribution bottles are installed to enable sorbent feed to either one or two rows of injectors at one time.

The 12 existing B&W circular burners at Edgewater were replaced with B&W XCL burners because of their ability to operate with the desired  $\text{NO}_x$  reduction while fitting within the space constraints of the Edgewater boiler.

Particulate control was a concern at Edgewater because LIMB increases particulate load by a factor of 2 to 3, and changes particulate properties such that the ash is more difficult to collect. The Edgewater ESP was retrofitted to the unit in 1982 and is substantially oversized for its present duty (the specific collection area is approximately  $600 \text{ ft}^2/1,000 \text{ acfm}$  under normal high load operating conditions), so no ESP modifications were made. LIMB ash collected in the ESP will contain approximately 30% free lime ( $\text{CaO}$ ) and is highly alkaline. The existing dry ash handling and storage system is being used, but a controlled amount of water is added to the ash as it is unloaded from the silo in order to hydrate the free lime for safe handling and disposal in an existing landfill.

#### HUMIDIFIER DESIGN

The Edgewater humidifier was designed to add sufficient water to the flue gas to bring the flue gas temperature to within a  $20^\circ\text{F}$  approach to the adiabatic saturation temperature. The actual temperature achieved in practice will be a trade-off between a low outlet temperature for higher sulfur removal and a higher temperature for improved operability. The humidifier was installed between the boiler and the ESP in a bypass arrangement (see Figure 2) on the boiler roof in a space previously occupied by a decommissioned ESP. A set of hoppers from the ESP was retained in the humidifier design to enable the removal of ash buildup in the humidifier, although initial operation has been with the ash hoppers covered to more resemble an in-duct system. The bypass arrangement was developed primarily in response to the untested reliability of the humidifier system at this time; the technology could possibly be installed in existing ductwork if this demonstration proves its reliability in production operation.

The flue gas humidifier is a horizontal chamber, 14 ft 7 in. x 14 ft 7 in. in cross section and 60 ft long, providing a roughly 2-second residence time at full load. The chamber is connected to the main flue by ductwork and a series of dampers. Turning vanes have been installed in the ductwork to distribute gas flow evenly through the chamber. With the bypass system, the flue gas can be diverted back to the main flue in case of humidifier malfunction or outage. Water is provided by a array of air/water spray atomizers located at the chamber entry. The array consists of 110 atomizers supported in 22 lances; each lance can be individually controlled and removed from the humidifier for repair without interrupting humidifier operation. Compressed air is used to provide the air to water weight ratio of up to 0.45 required for fine droplet formation in the spray pattern. The humidifier became operable in August 1988.

## START-UP EXPERIENCE AND RESULTS

The LIMB demonstration unit started up in July 1987 and operated through September 25, 1987. The early operating period was characterized by intermittent operation due to problems encountered with the sorbent feed system and the necessity of incorporating design changes to attain full flow capability. Once this had been achieved, a particulate removal problem was identified, relating to the high electrical resistivity of the ash produced with sorbent injection. This problem limited LIMB operation to 3 to 8 hours per injection period and made long-term, continuous operation impossible due to the need to maintain particulate emissions within the opacity limitations mandated by the Ohio EPA. Despite these time-consuming setbacks, a series of test runs were conducted over a 2-week period, and valuable operating information and experience was obtained. The emissions data gathered during this period show that the SO<sub>2</sub> removal capability of the LIMB technology exceeded the 50% removal goal originally set for the demonstration. The start-up experience is described in more detail in the following sections.

### OPERATING CONDITIONS

The coal fired during the 2-week test period was a high sulfur (2.5 to 3.0%) bituminous coal mined in Ohio. The sorbent was a calcitic hydrated lime obtained from a commercial vendor; an alternate modified sorbent, prepared by the same vendor by adding 1.5% by weight of calcium lignosulfonate to the lime during hydration, was also tested. Twelve tests were conducted under constant operating conditions at full boiler load. Due to lack of time to optimize the various injection parameters, one injection condition--use of only the lowest level of injectors at a set booster air flow rate and no injector tilt--was selected for most of the tests and held constant over the 2-week period. The only exceptions to this were one test using both the two lower levels and one test with the injection ports tilted down 15° from horizontal.

The overall performance of the total system--boiler, sorbent, injection, and emissions analyzers--was monitored by a computerized data acquisition system [B&W's Boiler Performance Diagnostics--System 140 (TM)]. In addition to its regular function of tracking normal boiler parameters, it was also programmed to perform a variety of calculations specific to the technology, such as Ca/S stoichiometry and momentum flux ratios, as well as to reduce the data according to specified procedures.

A continuous emission monitoring system was used to analyze several gases in the duct following the ESP. An extractive system was used to sample flue gas, which was then dried and split among the various analyzers which include SO<sub>2</sub>, NO, O<sub>2</sub>, CO<sub>2</sub>, CO, and total hydrocarbons. The filters in the sampling system were maintained at high (approximately 300° F) temperature to avoid capture of SO<sub>2</sub> in the sampling system.

Since the tests conducted were primarily directed at the SO<sub>2</sub> removal efficiency of the LIMB system as a function of the Ca/S stoichiometry, the calculations of particular importance were those associated with "inlet SO<sub>2</sub>," sorbent injection rate, and SO<sub>2</sub> outlet emissions. For continuous LIMB operation, inlet sulfur to the boiler is calculated by the data acquisition system from the input coal analysis and a series of heat and material balances derived from boiler measurements. This inlet sulfur calculation is the basis



for the sulfur term in the Ca/S stoichiometry ratio and is compared with the outlet  $\text{SO}_2$ , as measured by the continuous emission monitoring system, to calculate removal efficiency after appropriate conversions to a  $\text{lb}/10^6 \text{ Btu}$  basis. This removal efficiency is calculated as a 10-minute average by the data acquisition system. (Stoichiometry and numerous other parameters are likewise calculated as 10-minute averages.) This was considered the only practical method of determining  $\text{SO}_2$  removal efficiency during the demonstration, since continuous measurement of  $\text{SO}_2$  concentration in the lower furnace is impossible. However, during the 2-week test period described in this paper (when intermittent, short-duration tests were performed), the removal efficiencies reported were determined by comparing measured  $\text{SO}_2$  emissions before and after sorbent injection. This calculation method was considered to provide an even more accurate assessment of removal efficiency for these tests.

#### ELECTROSTATIC PRECIPITATOR OPERATION

At sorbent feed rates corresponding to a Ca/S ratio of 2.0 (10,000 to 13,000  $\text{lb/hr}$ ), operation was characterized by steady degradation of the ESP as indicated by an increase in stack opacity. Current/voltage relationships such as those shown in Figure 3 indicated that the high electrical resistivity of the ash was producing back corona in the ESP. The condition was so severe that all attempts to find an effective operating voltage were unsuccessful. Measurements of ash resistivity using an *in situ* point-plane probe showed that sorbent injection resulted in an increase from approximately  $3 \times 10^{10} \text{ ohm cm}$  to  $3 \times 10^{12} \text{ ohm cm}$ . As expected, mass train data showed that the particulate loading almost doubled, while particle size measurements indicated an increase in smaller particles during sorbent injection.

As a result of the degraded ESP performance, continuous sorbent injection was limited to 3 to 8 hours, depending on the feed rate. Typically, injection would be terminated when stack opacity would begin to rise rapidly into the 10 to 15% range in order to avoid exceeding the opacity standard of 20%. Upon termination of sorbent injection, stack opacity would gradually drift downward, and ESP electrical conditions would return to normal. Approximately 4 to 6 hours were usually required for return to the original condition where another sorbent injection test could be conducted.

One method of reducing LIMB ash resistivity and improving ESP performance during lime injection is to condition the flue gas. Past work has shown that both  $\text{SO}_3$  conditioning and humidification are effective means of lowering flyash resistivity. In the case of a LIMB ash, however,  $\text{SO}_3$  reacts with the lime-rich ash, requiring a very large excess of  $\text{SO}_3$ . A more practical method is water addition to the flue gas to increase humidity and reduce gas temperature, which in turn reduces resistivity to a level (about  $1 \times 10^{11} \text{ ohm cm}$ ) where normal ESP operation is possible. Figure 4 shows how the resistivity of LIMB ash varies with flue gas humidity and temperature (3).

#### LOW $\text{NO}_x$ OPERATION

A short series of tests were conducted with the low- $\text{NO}_x$  XCL burners prior to lime injection in order to allow comparison with data obtained on the circular burners they had replaced in 1986. As can be seen in Figure 5,  $\text{NO}_x$  emissions with the XCL burners achieved the project goal of  $0.5 \text{ lb}/10^6 \text{ Btu}$  over



the full operating range of the boiler (for the purposes of the project, the maximum continuous rating of the boiler's main steam flow is 700,000 lb/hr with a peak load of 770,000 lb/hr). This performance represents approximately 40-50% reduction when compared to data on the circular burners. Further burner tests are planned after optimization of sorbent injection. The data will be examined for interactions between SO<sub>2</sub> and NO<sub>x</sub> reduction, though none are expected at this time.

#### SULFUR DIOXIDE REMOVAL

Because of the impending boiler outage, the test period for determining SO<sub>2</sub> removal performance was limited to the 2 weeks prior to the outage. The first test week was devoted to operation with a commercial, hydrated calcitic lime sorbent and the second to operation with the same sorbent modified by the addition of calcium lignosulfonate. Bench-scale tests (4) had shown that addition of small amounts of calcium lignosulfonate during the hydration process resulted in a more reactive sorbent than normal hydrate. During this period, one or two tests of 1 to 5 hours duration (depending on stoichiometry) could be conducted during a day. Typically, the first few tests of each week were characterized by inconsistent sorbent feed and operational upsets which had an impact on performance, while the tests during the latter part of each week were characterized by steady-state operation. Once material handling problems were overcome, it appeared that a steady-state condition was reached very quickly after sorbent injection was begun and that this condition was representative of long-term operation. The results reported in this paper focus on the data considered to be most reliable during the 2-week period and do not include transitional periods or times when operational upsets were known to have occurred. It should be understood that the 2-week test period does not provide a definitive comparison between the normal commercial lime and the modified lime. However, the data are considered to be a reliable indicator of the type of results expected when more thorough testing resumes.

The overall relationship between SO<sub>2</sub> removal efficiency and Ca/S molar stoichiometry, as determined by the tests conducted, is shown in Figure 6. In this graph, each point represents the average value of all of the 10-minute averages taken during a test with a well-controlled feed rate. Each point represents data taken over a 1 to 5 hour period. The error limits shown for both SO<sub>2</sub> removal and stoichiometry reflect plus or minus twice the standard deviation calculated from the individual 10-minute averages making up each point.

A removal efficiency in the 55 to 60% range was obtained at a Ca/S stoichiometry of 2; the project goal of 50% removal appears obtainable at a stoichiometry of approximately 1.6. Future optimization of injection parameters is expected to improve the performance. Preliminary statistical analysis suggests that the modified hydrated lime exhibits slightly better performance. However, the nature of the tests conducted thus far does not support this as a strong conclusion concerning the relative merits of the two.

#### ON-GOING LIMB/HUMIDIFIER TESTS

Following completion of the bypass humidifier installation in August 1988, the LIMB/humidifier system operation and testing has been resumed. During initial operation, the humidifier has been operated with sufficient

humidification to allow the ESP to operate within the plant's opacity limitations. Humidifier outlet temperatures of roughly 250°F have been sufficient to enable continuous ESP operation with no detrimental effects from LIMB ash.

Initial testing of the system will focus on optimization of sorbent injection parameters (nozzle combinations, injection velocity, injection angle) to obtain maximum SO<sub>2</sub> removal in the boiler. Following this optimization program, the humidifier will be operated at progressively lower outlet temperatures to maximize the degree of humidification without jeopardizing operating reliability. Pilot tests indicate that SO<sub>2</sub> removal in the humidifier will increase as the relative humidity increases. Once these optimization activities are completed, the LIMB/humidifier system will be operated continuously in its optimum configuration and will follow boiler load routinely. Testing of the system in this mode will continue through February 1989.

### CONCLUSIONS

Operation and testing of the Edgewater LIMB system to date have provided positive indications of the viability of the technology. Short-term tests with hydrated lime were very encouraging as they showed SO<sub>2</sub> removal rates greater than 50% with stoichiometries less than 2. NO<sub>x</sub> emissions below 0.5 lb/10<sup>6</sup> Btu were shown to be achievable. Although ESP performance deteriorated during sorbent injection, early operation of the humidifier has indicated that this problem can be remedied with a modest degree of humidification.

The tests to date have not been sufficiently long to draw any definite conclusions about boiler operability under continuous sorbent injection. The initial ESP problems prevented operation for a time period sufficient to assess boiler effects over several soot blower cycles. More recent tests of longer duration have indicated, however, that boiler tube cleanliness can be maintained with increased soot blowing frequency.

Within the next few months, a clear picture should begin to emerge concerning the operability, effectiveness, and cost of the LIMB and flue gas humidifier technologies on a day-to-day basis.

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Readers more familiar with the metric system may use the following conversion factors:

| <u>From</u>            | <u>To</u>           | <u>Multiply By</u>   |
|------------------------|---------------------|----------------------|
| acfm                   | m <sup>3</sup> /min | 0.02832              |
| ft                     | m                   | 0.3048               |
| ft <sup>2</sup>        | m <sup>2</sup>      | 0.09290              |
| in.                    | m                   | 0.0254               |
| lb/hr                  | kg/h                | 0.45359              |
| lb/10 <sup>6</sup> Btu | ng/J                | 429.94               |
| ton                    | t                   | 0.90718              |
| μA/ft <sup>2</sup>     | ηA/cm <sup>2</sup>  | 1.07639              |
| °F                     | °C                  | °C = (5/9) (°F - 32) |

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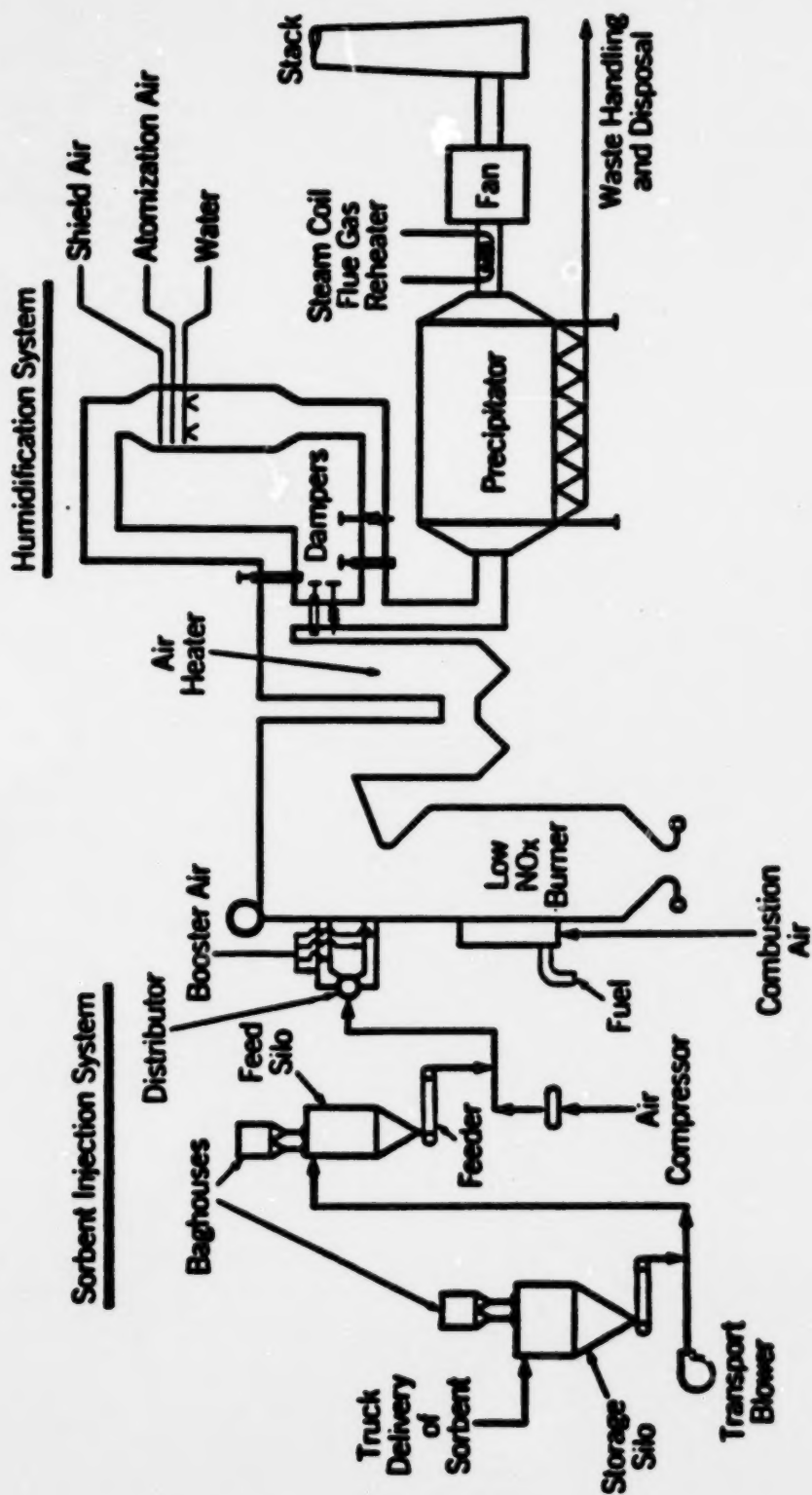


Figure 1. LIMB process flow diagram.



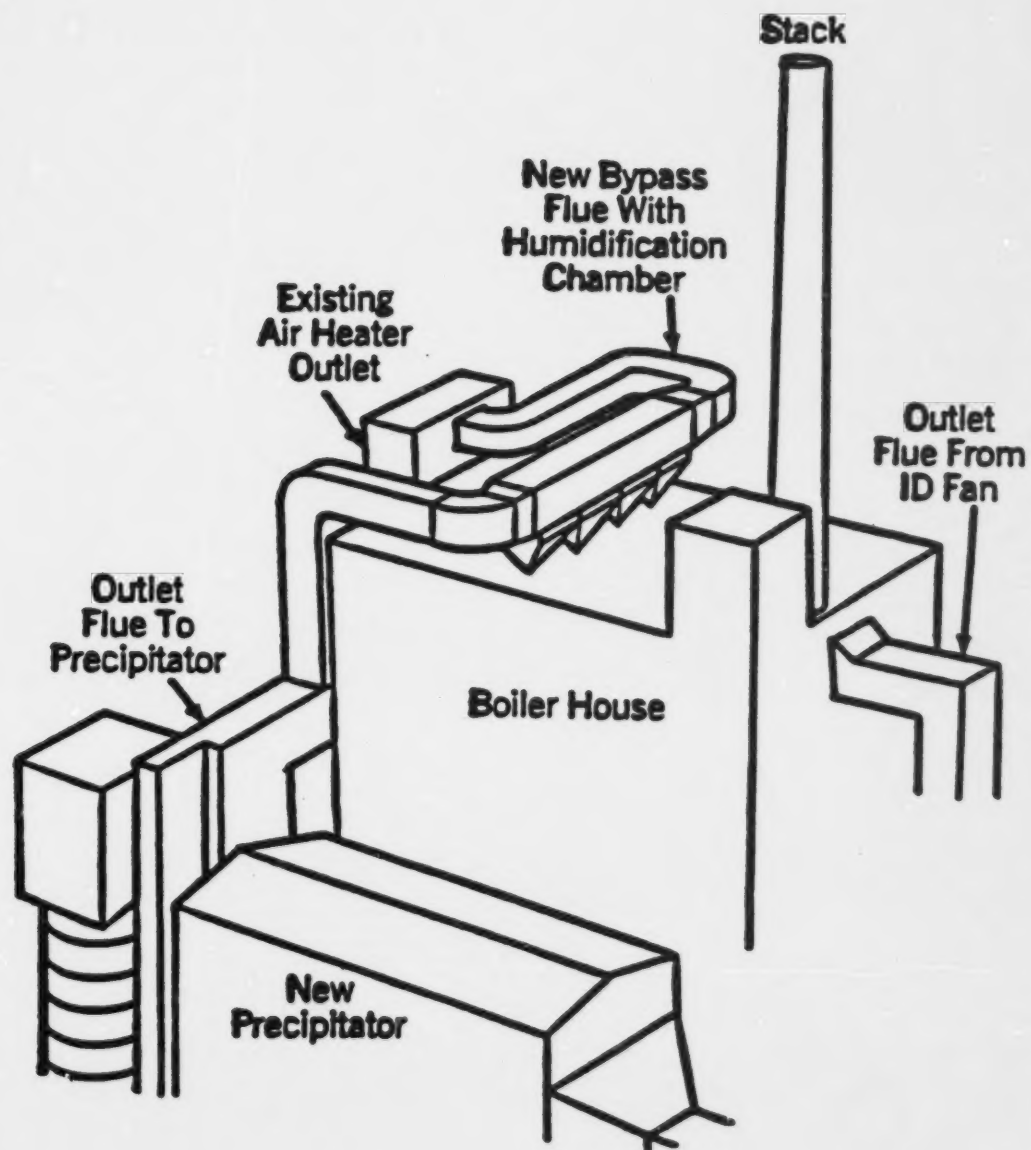


**Figure 2. Bypass humidification chamber.**

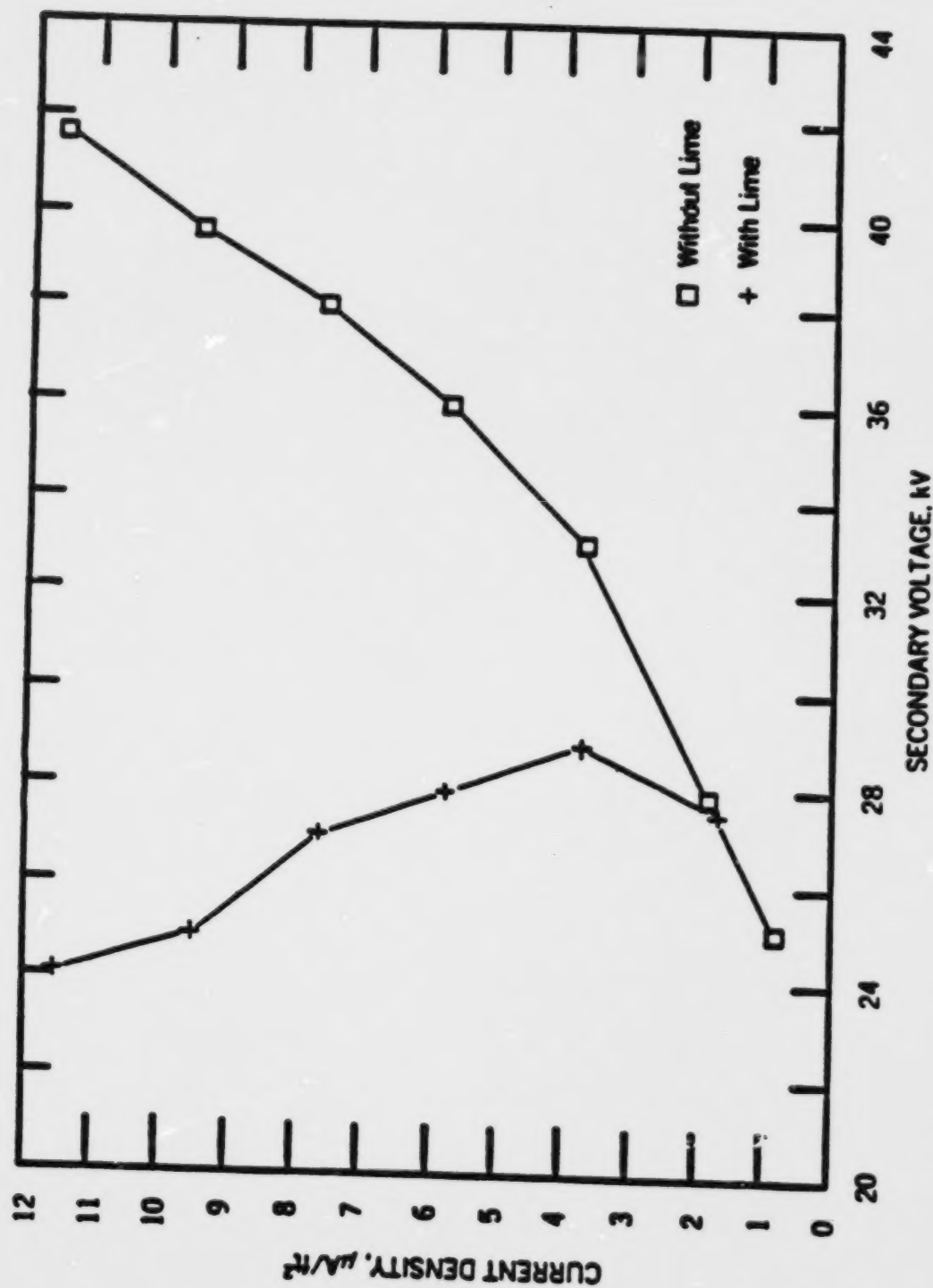


Figure 3. ESP current/voltage relationships with and without lime injection.

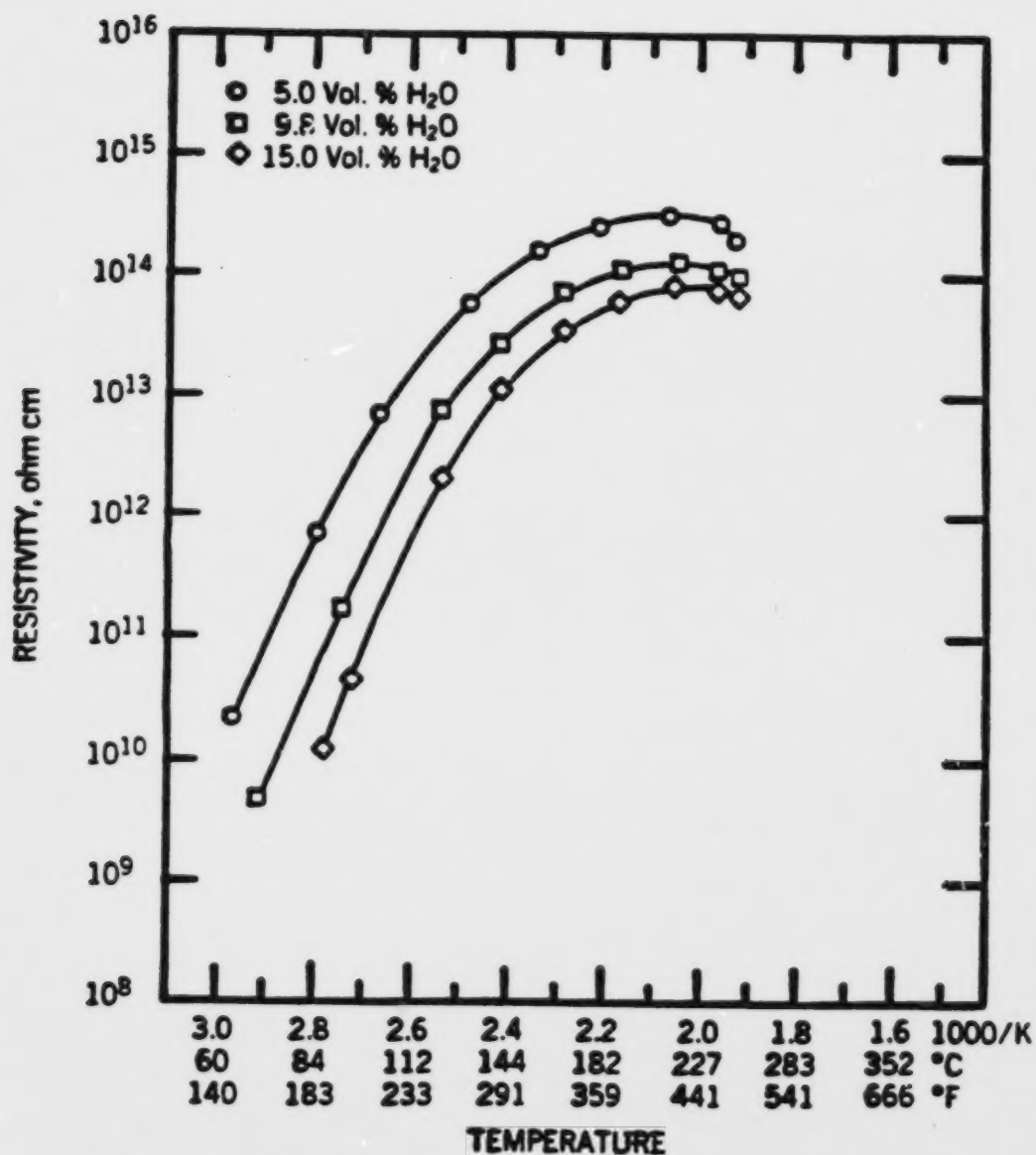


Figure 4. LIMB ash resistivity as a function of temperature and humidity.

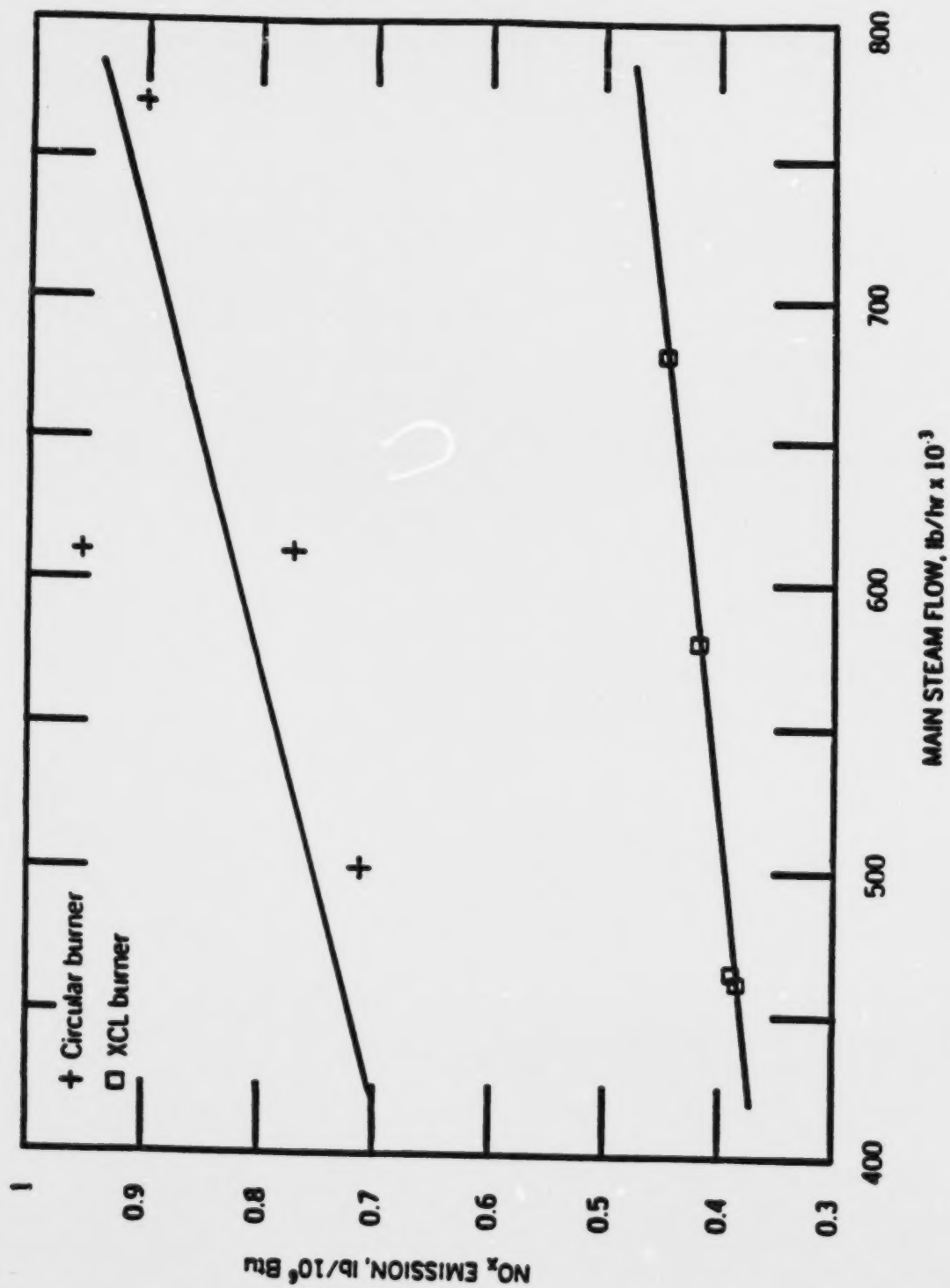


Figure 5. NO<sub>x</sub> emission as a function of boiler load.



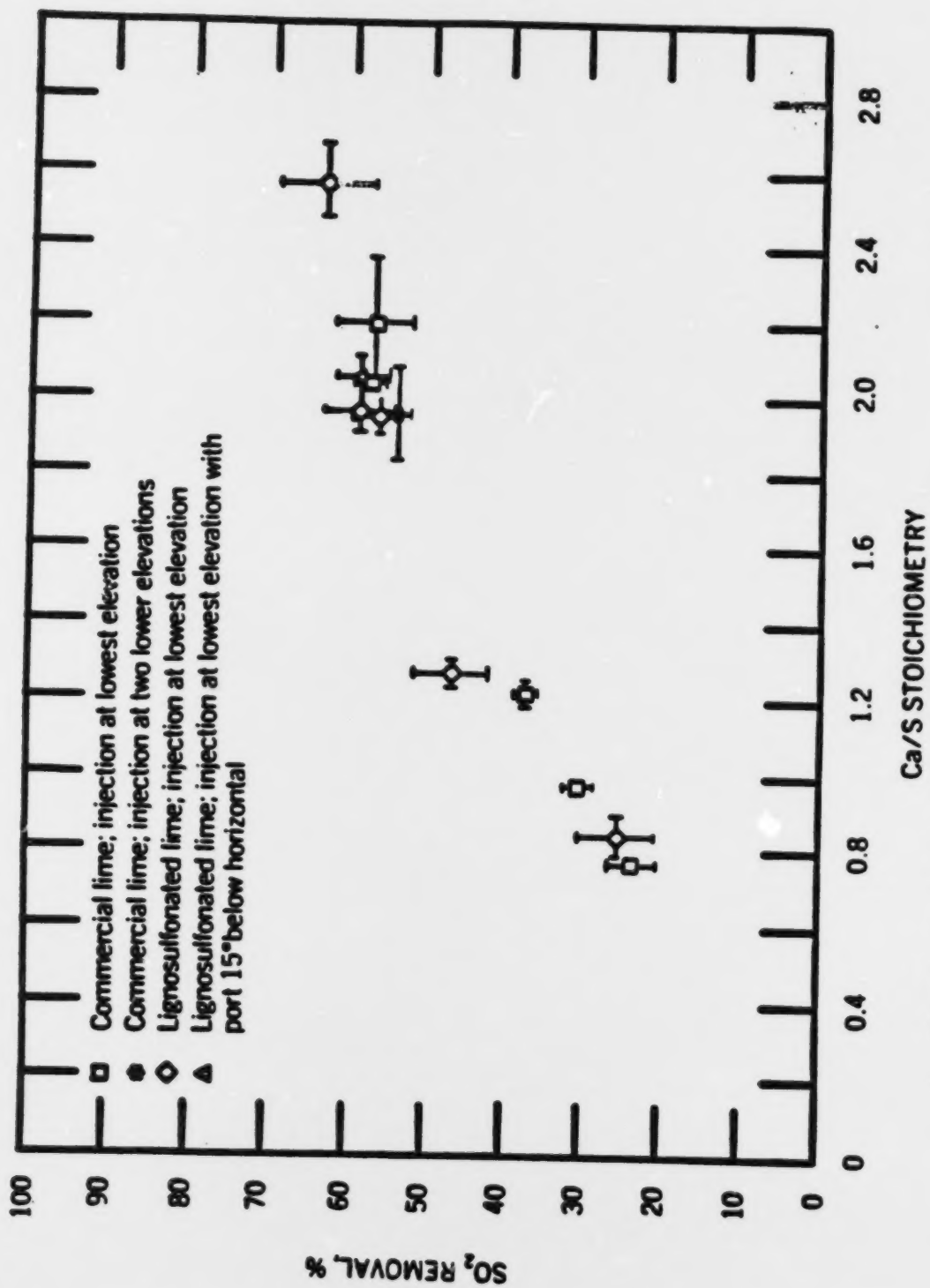


**Figure 6. Sulfur dioxide removal efficiency as a function of calcium/sulfur stoichiometry.**

PROTOTYPE EVALUATION OF SORBENT INJECTION WITH HUMIDIFICATION

by: G. C. England, B. A. Folsom, R. Payne, and T. M. Sommer  
Energy and Environmental Research Corporation  
Irvine, California 92718

M. W. McElroy  
Electric Power Research Institute  
Palo Alto, California 94303

P. J. Chappell  
Air and Energy Engineering Research Laboratory  
U. S. Environmental Protection Agency  
Research Triangle Park, North Carolina 27711

I. A. Huffman  
Richmond Power and Light  
Richmond, Indiana 47313

ABSTRACT

This paper describes a test program being conducted on a 61 MW coal fired utility boiler to determine factors controlling SO<sub>2</sub> emissions reduction by injection of calcium-based sorbents into the furnace. A flexible sorbent injection system has been designed and installed on Unit 2 of Richmond Power and Light's Whitewater Valley Generating Station located in Richmond, Indiana. Unit 2 is tangentially fired and has a cold side electrostatic precipitator (ESP). Tests are currently in progress injecting a wide range of sorbents -- including hydrated lime, limestone, dolomitic hydrated lime, and a promoted sorbent -- into the upper furnace of the boiler. Preliminary results during short-term tests with hydrated lime show that SO<sub>2</sub> emissions can be reduced by 25-40% at Ca/S = 2.0. Further optimization of the injection system is planned to define maximum SO<sub>2</sub> capture compared to predicted levels of 45% at Ca/S = 2.0 and full boiler load.

A flue gas humidification system was designed and installed. Humidification has proven very effective in restoring ESP electrical performance during short term operation at flue gas temperatures of 240-270°F (116-132°C) in the ESP. Pre-existing stack opacity levels have been maintained during sorbent injection, although instances of higher opacity have been observed. Additional characterization of ESP performance with sorbent injection and humidification is planned, including resistivity and particle size measurements.

## INTRODUCTION

Energy and Environmental Research Corporation (EER) is conducting a prototype evaluation of sorbent injection technology for SO<sub>2</sub> emissions control on a 61 MW tangentially-fired pulverized coal utility boiler. Sorbent injection into the boiler furnace reduces SO<sub>2</sub> emissions by reaction of calcium-based sorbents with SO<sub>2</sub> in the furnace gases to form calcium sulfate, a solid product which is then removed in the existing dust collector. The objective of this project is to evaluate the effects of sorbent injection process parameters on SO<sub>2</sub> removal and on boiler/ESP performance under realistic utility operating conditions. The process parameters include sorbent type, injection condition, and injection configuration. To this end, a very flexible sorbent injection system has been installed at the host boiler to enable operation over a wide range of injection conditions. The project will culminate in a 15 day test in which the sorbent injection system will operate 24 hours per day under optimized process conditions, over the normal duty cycle of the boiler.

Sorbent injection degrades ESP performance due to increased dust resistivity, increased fine particulate matter, reduced SO<sub>3</sub> concentration, and increased total dust loading. A flue gas humidification system has been installed in the duct between the air heater and the ESP for the primary purpose of counteracting these effects. Evaporative humidification is thought to greatly reduce dust resistivity while also slightly increasing the specific collection area (SCA) of the ESP by reducing the gas volume. At very low flue gas temperatures approaching the water saturation temperature, incremental SO<sub>2</sub> removal is also possible, but there are currently no plans to test this effect.

This project is jointly sponsored by the U.S. Environmental Protection Agency (EPA) and the Electric Power Research Institute (EPRI), and is one of three projects currently underway on full scale utility boilers in the U.S. The emphasis of this project is on evaluating a wide range of process parameters during relatively short periods of operation in order to enhance understanding of the process, as opposed to a demonstration of sorbent injection over an extended time period under optimum sorbent injection conditions. Specifically, the project is intended to provide information on process design, performance, and plant operational impacts for tangentially-fired boilers that will support the design of the last of these projects -- the Yorktown sorbent injection demonstration on a 180 MW tangentially-fired boiler (1).

This paper describes the host boiler and the design of the sorbent injection and humidification systems. Start-up of the sorbent injection/ humidification system has just been completed and testing has been initiated. Preliminary results, including SO<sub>2</sub> removal, boiler impacts, and ESP performance, are also discussed.

## BACKGROUND

The injection of dry sorbents into the furnace of a coal fired boiler for SO<sub>2</sub> emissions control may be a cost effective alternative to flue gas scrubbing in instances where moderate (e.g., <60%) SO<sub>2</sub> reduction is required. The efficiency of SO<sub>2</sub> reduction is controlled by three mechanisms:

- sorbent activation -- upon injection into the hot furnace gases the sorbent calcines to CaO under conditions promoting high particle surface area and reactivity;
- sorbent deactivation -- occurs at high temperatures where sintering reactions reduce particle surface area and reactivity; and



- sulfation -- reaction between  $\text{SO}_2$  and  $\text{CaO}$  occurs within an optimum temperature range, producing a solid product ( $\text{CaSO}_4$ ) which can be removed in the dust collector.

The optimum process conditions for injecting sorbents have been studied extensively in recent years. The optimum gas temperature for injection of the sorbent into the furnace has been established to be approximately 2250°F (2,3). Injection at temperatures above optimum leads to deactivation of the sorbent particles and decomposition of the reaction product. The optimum injection temperature generally occurs in the upper furnace of coal fired boilers near the entrance to the convection section at full design load. Sulfation proceeds at significant rates down to gas temperatures of approximately 1550-1600°F, generally somewhere near the exit of the superheater. Injection at temperatures below optimum reduces the residence time available within these temperature boundaries, leading to reduced  $\text{SO}_2$  removal. The characteristics of the sorbent also affect  $\text{SO}_2$  removal; however, optimum process conditions for furnace sorbent injection are very similar for all calcium-based sorbents. High calcium hydrated limes generally have a higher reactivity than limestones (3), although some limestones have been shown to be as reactive as some of the less reactive hydrated limes. Dolomitic hydrates generally are more reactive on the basis of  $\text{Ca/S}$ , but are approximately equal to hydrated limes on an equal mass basis. Sorbent reactivity does not correlate well with physical and chemical characteristics of the raw sorbent; however, sorbent reactivity generally increases as sorbent particle size decreases.

#### HOST BOILER

The host boiler for the project (Figure 1) is a 61 MW tangentially fired boiler built in 1972 by Combustion Engineering (CE). Unit 2 of Whitewater Valley Generating Station is owned and operated by Richmond Power and Light (RP&L), a municipal utility in Richmond, Indiana. Table 1 summarizes the design features of the unit. This unit fires a 2.5% sulfur Indiana bituminous coal.  $\text{SO}_2$  emissions are limited to 6.0 lb/10<sup>6</sup> BTU on a 30-day rolling average based on coal sulfur content. The firing system employs three Raymond-CE bowl mills which each supplies one elevation of coal nozzles at each corner of the furnace. The burners were modified in 1980 to incorporate CE's first generation Concentric Firing System (CFS) (4). The modification consists of diverting a portion of the combustion air away from the main firing circle toward the walls of the furnace, creating a low excess air fireball and effectively staging the combustion process. The burners do not have tilt capability. The furnace is equipped with 24 wallblowers to control wall deposits, 12 above the burners and 12 below. The combustion products enter platen and pendant superheaters at approximately 1900°F. The superheater section is equipped with four retractable sootblowers. Attemperation spray is added to the steam circuit between the superheater sections for steam temperature control. Typical of medium pressure boilers, a convective boiler section with two retractable sootblowers is positioned between the superheater and economizer. The economizer is a finned tube design with no sootblowers. The air heater is a conventional Lungstrum horizontal axis regenerative unit.

The unit is equipped with a Lodge-Cottrell cold-side ESP with a nominal design SCA of 198 ft<sup>2</sup>/1000 acfm. The unit has four separately energized electrical fields in series, and the transformer-rectifier (T-R) set controllers are of the saturable reactor type. The flue gases flow through a relatively long duct from the air heater to the ESP inlet. There are several bends. The ESP design utilizes mechanical rappers on the discharge and collecting electrodes. The normal particulate emissions requirements are a maximum of 20% opacity and 0.1 gr/dscf.

Bottom ash is sluiced to a dewatering bin and trucked to an off-site landfill. Fly ash is handled with a conventional dry vacuum system utilizing a water



exhauster. The fly ash is transferred to a temporary storage silo, unloaded to trucks via a dustless unloader, and trucked to a landfill.

### SORBENT INJECTION PROCESS DESIGN

The key elements of the design procedure are: (1) isothermal physical flow modeling of the boiler furnace to determine gas flow field characteristics and to evaluate injector configurations for optimum mixing between the sorbent and furnace gases; and (2) mathematical heat transfer modeling based on the flow field to determine the temperature field within the furnace and to evaluate impacts on boiler performance. The heat transfer model predictions were complemented with field measurements of furnace gas temperature, velocity, and species concentrations to improve the accuracy of the predictions. The anticipated performance of the sorbents was predicted by coupling the results of experimental characterization of sorbent reactivity and the predicted thermal history experienced by the sorbent, in a model of sorbent calcination and sulfation.

The furnace flow field of the RP&L boiler was characterized in a 1/12 scale physical model of the unit. The approach to isothermal modeling of the unit is described in detail elsewhere (5). The flow field within the region of the furnace above the burners is characterized by a strongly swirling flow, rotating counterclockwise in plan view, with high velocities near the walls and relatively low velocities in the center. The gas flow in the region near the nose of the boiler, where the gases turn to enter the superheater and convective passes, is very complex. The center of this region contains a large weak recirculation zone. Most of the gas flow turns abruptly over the nose on the back wall, or travels along the walls and enters the superheaters on the right side of the boiler (as viewed from the front). Field measurements confirmed the overall structure of the flow field.

The mean axial temperature profile in the RP&L boiler is shown in Figure 2. The results of the heat transfer model are compared with field measurements for both nominal full load (64 MW) and low load (40 MW) operating conditions. The results show that the optimum temperature for sorbent injection is located approximately at the nose at full load and approximately 2 m above the burner region at low load.

Since the flow field at the optimum injection temperature zone is very three-dimensional, optimum mixing between the injected sorbent and the furnace gases was predicted to require placement of injectors at the locations illustrated in Figure 3. The diameter of the injectors and injection velocity were selected to provide penetration into the main gas flow, but not into the central recirculation zone. Since most of the gas mass flow in the nose region is concentrated near the walls, a relatively low injection velocity (140 ft/sec) was predicted to provide optimum mixing at full load. Low load injectors are located in the existing furnace observation ports above the burner belt, oriented counter to the rotation of the furnace gases. Optimum injection velocity for the lower injectors was 330 ft/sec.

### HUMIDIFICATION SYSTEM PROCESS DESIGN

Humidification involves injection of water as a fine spray into the flue gases downstream of the air heater. Because the spent sorbent in the flue gases will contain a relatively high fraction of CaO, formation of cementitious deposits in the duct and ESP may occur if the sprays impinge on and wet the walls, or if evaporation is incomplete at the ESP entrance. Figure 4 illustrates the duct between the air heater and the ESP on the RP&L boiler. The residence time in the initial vertical section of the duct is approximately 0.75 sec at full load, while the total residence time to the ESP inlet is approximately 2.0 sec.

The water sprays are installed in the duct as near to the air heater as practical to maximize evaporation prior to the ESP. The residence time from this injection location to the first bend is 0.45 second. Field measurements indicated that the flow leaving the elbow immediately downstream of the air heater was very complex due to flow separation from the inside radius of the bend and the semicircular shape of the air heater gas outlet. This resulted in a non-uniform velocity profile and recirculation at the humidification plane. The elbow at the air heater outlet was redesigned to incorporate turning vanes, providing a more uniform velocity profile at the humidification plane and reducing the possibility of deposit formation due to recirculation.

The initial design goal was to achieve a 30°F approach to the adiabatic saturation temperature. The performance of the spray nozzles is a key variable controlling droplet evaporation and wall wetting, and therefore the actual approach to saturation which can be achieved. One- and two-dimensional mathematical models of droplet evaporation and trajectories were utilized to define spray nozzle performance requirements, design the nozzle array, and estimate the overall performance of the system (6). The calculations indicated that very fine sprays with Sauter mean diameter (SMD) below approximately 20  $\mu\text{m}$  would be required to achieve complete evaporation prior to the ESP at 30°F approach to saturation, and that large droplets would probably impact on the wall of the duct. Relaxing the specifications slightly indicated that larger droplets can be tolerated since droplet lifetimes are significantly reduced as the temperature above saturation increases. Since atomizers with SMD of 20  $\mu\text{m}$  in the capacity range of interest were not offered commercially, the nozzle performance specifications were modified to be SMD below 30  $\mu\text{m}$  with a minimum percentage of large droplets.

The spray characteristics produced by a range of atomizers were measured. The atomizers selected for testing consisted of commercially available twin-fluid and pressure-atomized designs. Twin-fluid designs generally have better turndown characteristics, but require a source of high pressure (typically 100 psig) air. The air requirement can be a significant auxiliary power requirement in terms of compressor capacity. Pressure atomizers are therefore attractive because there is no compressed air or steam required for atomization, but generally spray quality deteriorates with turndown and increasing nozzle capacity. Two design concepts were evaluated: an array of twin-fluid nozzles with approximately 2 gpm capacity each; and an array of smaller pressure nozzles with 0.3 gpm capacity each. Spray angles were below 20 degrees to minimize spray overlap. For the twin-fluid nozzles, maximum air/water mass ratio was 0.3 to limit compressed air requirements.

The SMD and fraction of droplets above 112  $\mu\text{m}$  are presented in Figure 5 for each of the atomizers tested. The figure shows that only one of the twin-fluid designs approached (but did not meet) the performance requirements. The pressure atomizers appeared to produce similar spray quality based on measurements made at the nozzle centerline; however, the periphery of the spray contained a large number of large droplets.

Nozzle 7 was selected for the final design. The optimized design of the nozzle array incorporated 31 nozzles, with a minimum nozzle-wall spacing of 1.4 ft and minimum nozzle-nozzle spacing of 1.37 ft to minimize the possibility of wall wetting and achieve uniform humidification. Performance estimates were made for the optimized design using mathematical models (6) which indicated that the closest approach to saturation possible while maintaining complete evaporation at the ESP is 70°F.

#### WASTE DISPOSAL

The fly ash generated during sorbent injection is enriched with spent sorbent (primarily  $\text{CaO}$  and  $\text{CaSO}_4$ ). The properties of the fly ash/spent sorbent mixture are

different from those of normal fly ash; therefore, modifications to handling and disposal procedures are necessary. Relative to normal fly ash, fly ash/spent sorbent mixtures exhibit increased pozzolanic activity upon addition of water, and the leachate from dry disposal sites can have pH levels approaching 12 and generally has increased concentrations of suspended solids and sulfates. The State of Indiana is currently revising regulations concerning disposal of solid waste. Since the properties of fly ash/spent sorbent mixtures could impact the future disposition of the current fly ash disposal site, the fly ash/spent sorbent generated during the program is being taken to a nearby sanitary landfill as a short term solution.

#### DESIGN SPECIFICATIONS

The design specifications for the sorbent injection and humidification systems were established based on the need for operation over a wide range of process conditions. The capacity of the sorbent feeding system was selected to feed hydrated lime, limestone, and dolomitic hydrated lime at Ca/S ranging from 1.0 to 3.0 with medium sulfur (2.5%) and high sulfur (4.5%) coals and at boiler loads ranging from 40 to 64 MW. The sorbent transport/injection system is designed to provide injection velocities from approximately 100-300 ft/sec. The capacity of the injection air supply was limited to 5% of the total air supplied to the boiler, which is considered the practical limit by which excess air may be increased in the boiler without excessive impact on thermal performance or generating capacity. The system was designed to allow injection with the nominal sorbent (hydrated lime) and coal (2.5% sulfur) on a 24 hour per day basis.

The capacity of the humidification system was sized to cool the flue gases to within 30°F of the adiabatic saturation temperature. While significant enhancement of ESP performance was anticipated at higher temperatures and calculations indicated that severe deposits were likely to form at such a close approach to saturation, the capacity was selected so that optimization of the system for SO<sub>2</sub> capture could be performed later.

#### ENGINEERING DESIGN

Figure 6 shows an overall process flow diagram for the plant including the sorbent injection and humidification systems. Sorbent is conveyed pneumatically from the storage/feeding system and injected directly into the boiler. Circulating water is pumped to the humidification nozzles and sprayed into the flue gas duct.

Sorbent is delivered in trucks and unloaded pneumatically to a silo. Discharge from the hopper is aided by air slides installed on the hopper slope. The silo vents are equipped with two bag filters which release air from silo filling and the air slides. A rotary valve meters the sorbent into a weigh-auger feeder. The feeder measures sorbent flow rate using a fixed-speed screw feeder which is mounted on a weighing mechanism. Feed rate is determined by multiplying screw speed by the weight of sorbent in the screw. The weigh auger feeder discharges into a solids pump (Fuller-Kinyon pump), which injects the sorbent into the elevated-pressure pneumatic conveying line. The solids pump is a high speed screw feeder which forces the solids through a check valve. The check valve prevents backflow of air from the conveying line. The weigh auger feeder is extremely sensitive to pressurization and must be maintained at atmospheric pressure; therefore, any air leaking back from the conveying line must be fully vented. A vent system was designed and installed at the inlet and outlet of the feeder to relieve air pressure originating at the feeder inlet or outlet. The sorbent is pneumatically conveyed from the solids pump to a splitter which distributes the sorbent to each sorbent injector. Additional injection air is mixed with the sorbent at the injectors to provide the air flow needed for the



proper injection velocity. The transport and injection air is supplied by two positive displacement blowers.

The current sorbent storage/feed/delivery system is a result of several modifications to the original design needed to overcome inherent difficulties feeding hydrated lime. The design of the system is intended to be extremely flexible, accommodating a wide range of sorbent types, feed rates, and injection configurations. High-reactivity sorbents have proven very difficult to feed due to fine particle size, low bulk density, and low bulk permeability. Table 2 highlights the original and modified system design. The most significant modifications to the sorbent system were:

- Removal of a vibrating discharger from the silo, which was causing compaction and uneven discharge from the silo when feeding hydrated lime. The vibrating discharger was replaced with the 60 degree straight hopper and air slides. A rotary valve was added to control silo discharge rate control.
- Replacement of a rotary valve with a high speed solids pump (Fuller-Kinyon pump) for transfer of the sorbent into the conveying line. Air leakage back through the rotary valve exceeded the capacity of the existing air vent system, pressurizing the feeder, disrupting feeder operation, and preventing even flow of sorbent. The capacity of the vent system was also increased.
- Separation of the sorbent conveying system from the injection air system. In the original design, the conveying line carried all of the injection air in an 8-in spiral wound steel pipe. The relatively large line size was necessary to accommodate the wide range of planned injection configurations. Surging sorbent flow in the conveying system was traced to material dropout in the conveying lines, even at transport velocities well above recommended minimum levels. The conveying and injection air systems were later separated, providing a separate 5-in rubber sorbent transport line. The existing steel line was retained for auxiliary injection air, which is added at the sorbent injectors to provide the required injection velocity.
- Modifications to the solids pump and the weigh-auger feeder drive speeds to increase system capacity. Equipment vendors underestimated the feed capacity of their equipment with hydrated lime.

Acceptable system performance was achieved after the above modifications were made.

The design of the humidification system is relatively straightforward. Circulating water is pumped through 60-mesh strainers to the nozzle array. Atomization air is supplied by three 200 hp rotary air compressors. Humidification water flow rate can be controlled either manually or automatically to control ESP exit gas temperature. The 31 spray nozzles are mounted on 6 lances. Three lances are inserted from each side of the duct, forming three rows of nozzles. Due to the concern over potential deposit formation in the humidification duct, the duct is extensively instrumented and monitored. Eighty-four thermocouples are installed on the duct walls to monitor wall wetting, and many ports were added along the duct for visual inspection of the duct walls and spray nozzles. The hanger rods supporting the duct were also instrumented with load cells to detect larger deposits.



## DESIGN PERFORMANCE ESTIMATES

The expected SO<sub>2</sub> removal achievable via sorbent injection was calculated based on the procedures described earlier. Initially, four sorbents were selected for evaluation during the test program which represent the full range of commonly available calcium based sorbents. The reactivity of the selected sorbents was determined in a bench-scale test furnace firing the host site coal under conditions approximating those in the host boiler. A hydrated lime treated with lignosulfonate to enhance reactivity (7) will also be tested. Table 3 compares the properties of the sorbents and results of the bench-scale tests. Table 4 presents estimated SO<sub>2</sub> removal in the RP&L boiler for four of the selected sorbents, based on sulfation model calculations. Calculations were performed for optimum and non-optimum injection conditions. At full load, the Marblehead hydrated lime is expected decrease SO<sub>2</sub> by 45% with the optimum injection (nose elevation) configuration. Injection of sorbent into the lower injection elevation (10 m), where the temperature is above optimum, is expected to result in 39% SO<sub>2</sub> removal. The highest SO<sub>2</sub> removal (65%) at full load, Ca/S = 2.0, and optimum injection conditions is expected with the Corson dolomitic hydrate. The lowest SO<sub>2</sub> removal (33%) under similar conditions is expected with the limestone.

Humidification system performance was estimated based on the final nozzle selection and design configuration. The performance estimates indicated that complete evaporation of the water sprays at 30°F approach to saturation would not be possible using the current nozzles. The lowest temperature achievable with complete evaporation was estimated at 190°F, corresponding to a 70°F approach to saturation. While this is not expected to produce significant incremental SO<sub>2</sub> removal, available literature suggests significant ESP performance improvement should be achieved.

## RESULTS

Preliminary data regarding system performance have been obtained during recently completed start-up tests conducted on the RP&L boiler. The start-up tests have been oriented toward demonstration of equipment performance and definition of acceptable operating conditions rather than optimization of system performance. Preliminary SO<sub>2</sub> removal data, based on SO<sub>2</sub> measurements made after the ESP, are presented in Figure 7. These data were obtained at full load with Marblehead hydrated lime. The sorbent was injected through the 8 injectors located at the upper injection elevation at 140 ft/sec with Ca/S=2.0. The sulfur capture and calcium utilization both peak in the range of 50 MW, but there is substantial data scatter. These factors are probably due to the gas temperatures in the upper furnace and convective pass. Suction pyrometer temperature measurements made during sorbent injection are about 150-200°F hotter than similar measurements at the same location made during baseline tests. The baseline measurements were used for the sorbent injection system design and performance projections. The sulfur capture is lower than predicted, possibly due to the higher temperatures. Also, the temperatures vary with furnace and convective pass deposits which are affected by sootblowing. The data scatter in Figure 7 is probably due to these sootblowing effects. Additional tests are planned to determine the reasons for the temperature increases and to optimize sootblowing and sorbent injection parameters for the actual host boiler conditions.

Humidification has proven to be extremely effective as an ESP performance enhancement during short term operation. Steady-state operation with sorbent injection could not be achieved without humidification during these tests because of high opacity. Observation of ESP secondary voltage indicated that excessive sparking occurred when sorbent was injected without humidification. The ESP controls would adjust the secondary voltages downward to compensate for excessive

sparking. This was particularly evident in the inlet field of the ESP. This would result in high opacity after a very short period of time.

The major benefit of humidification is that it allows the ESP to operate at increased power levels without back corona by reducing fly ash resistivity. Figure 8 illustrates the effect of reducing ESP gas temperature (by increasing humidification) on stack opacity during short-term tests conducted during start-up. The initial short term data indicate that opacity decreases as ESP temperature is reduced, and that opacity can be reduced below the required normal limit of 20% with ESP temperature of 270-280°F. ESP performance is generally better at low load, due to higher SCA, allowing higher sorbent injection rates.

Some brief tests were conducted over a 4 hour period at full load, Ca/S=2.0 (approximately 7500 lb/hr), and ESP temperature of 240°F. Opacity was maintained between 8-12% during the test; however, increased rate of deposit buildup was observed in portions of the duct and humidification was subsequently reduced. The ESP T-R sets were operating at or near the current limit during the test. Most of the subsequent testing has been conducted at higher temperatures with 15-25% opacity.

While the longest period of continuous operation to-date is only 29 hours, no long term adverse effects on ESP performance have been observed. Inspection of the duct and ESP after over 300 hours of sorbent injection with humidification showed no permanent buildup of material on the ESP electrodes, and only minor deposits in the duct immediately downstream of the spray nozzles. Heavier deposits were found in the first horizontal duct section downstream of the humidification sprays, which are believed to have formed during periods of operation at low ESP temperature. Some material has also collected at the bottom of the vertical duct, primarily deposits which formed on the nozzles and were subsequently detached. The data also indicate that boiler fouling will impact ESP performance due to changes in air heater exit temperature. This will affect the amount of water injection necessary to maintain constant ESP temperature.

Sorbent injection at Ca/S = 2.0 has an immediate effect on boiler fouling, particularly in the primary superheater. Figure 9 shows boiler gas temperatures during sorbent injection at full load. The air heater gas outlet temperature increases from approximately 325°F without sorbent injection to 355°F with injection at Ca/S = 2.0. As the superheater fouls, attenuation water flow rate decreases to maintain steam temperature. As shown in the figure, the sootblowers are effective in removing the deposits formed during sorbent injection. The sootblowers in the superheater and boiler must be operated almost continuously to maintain steam temperature and exit gas temperature within acceptable limits. At present, the maximum sorbent injection rate appears to be limited by high air heater gas inlet temperature due to boiler fouling. Current testing is focusing on optimization of sootblowing to minimize fouling and to define the maximum practical sorbent injection rate.

#### CONCLUSIONS

Preliminary data show that SO<sub>2</sub> removals up to approximately 40% were achievable at full boiler load with hydrated lime at Ca/S = 2.0, without optimization of injection parameters. The results also suggest that humidification will enhance ESP performance during short term sorbent injection. However, long term performance has not been demonstrated, and the system was not operated at close approach to saturation to establish the lowest achievable opacity when injecting sorbent. Also, boiler fouling due to sorbent injection resulted in increased flue gas exit temperatures. Additional characterization of the impact of system operation and sootblowing cycles will be necessary before boiler impacts and

system performance can be assessed definitively. Additional tests are currently in progress.

#### ACKNOWLEDGEMENT

This work was funded under EPRI Contract RP-2786-2 and EPA Contract 68-02-4253. The authors wish to acknowledge the efforts of E. Mecchia of EER for his efforts during the design and start-up of the LIMB system. The authors also wish to acknowledge the outstanding cooperation and support from Richmond Power and Light, notably I Huffman, R. Ray, S. Rexrode, I. Rowe, and D. Straight.

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#### METRIC EQUIVALENTS

|                                |                                               |                |                                             |
|--------------------------------|-----------------------------------------------|----------------|---------------------------------------------|
|                                | $^{\circ}\text{F} - 32$                       |                |                                             |
| $^{\circ}\text{C}$             | $= \frac{\quad}{1.8}$                         | kW             | $= \text{hp} \times 0.7457$                 |
| kg                             | $= \text{lb} \times 0.454$                    | l/min          | $= \text{gpm} \times 3.785$                 |
| m                              | $= \text{ft} \times 0.305$                    | $\text{g/m}^3$ | $= \text{gr/ft}^3 \times 2.29$              |
| mm                             | $= \text{inch} \times 25.4$                   | W              | $= \text{Btu/hr} \times 0.293$              |
| $\text{m}^2$                   | $= \text{ft}^2 \times 0.0929$                 | Pa             | $= \text{psig} \times 6894.8$               |
| $\text{m}^3$                   | $= \text{ft}^3 \times 0.0283$                 | $\text{g/MJ}$  | $= \text{lb}/10^6 \text{ Btu} \times 0.430$ |
| $\text{m}^2/1000 \text{ acmm}$ | $= \text{ft}^2/1000 \text{ acfm} \times 3.28$ |                |                                             |

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.



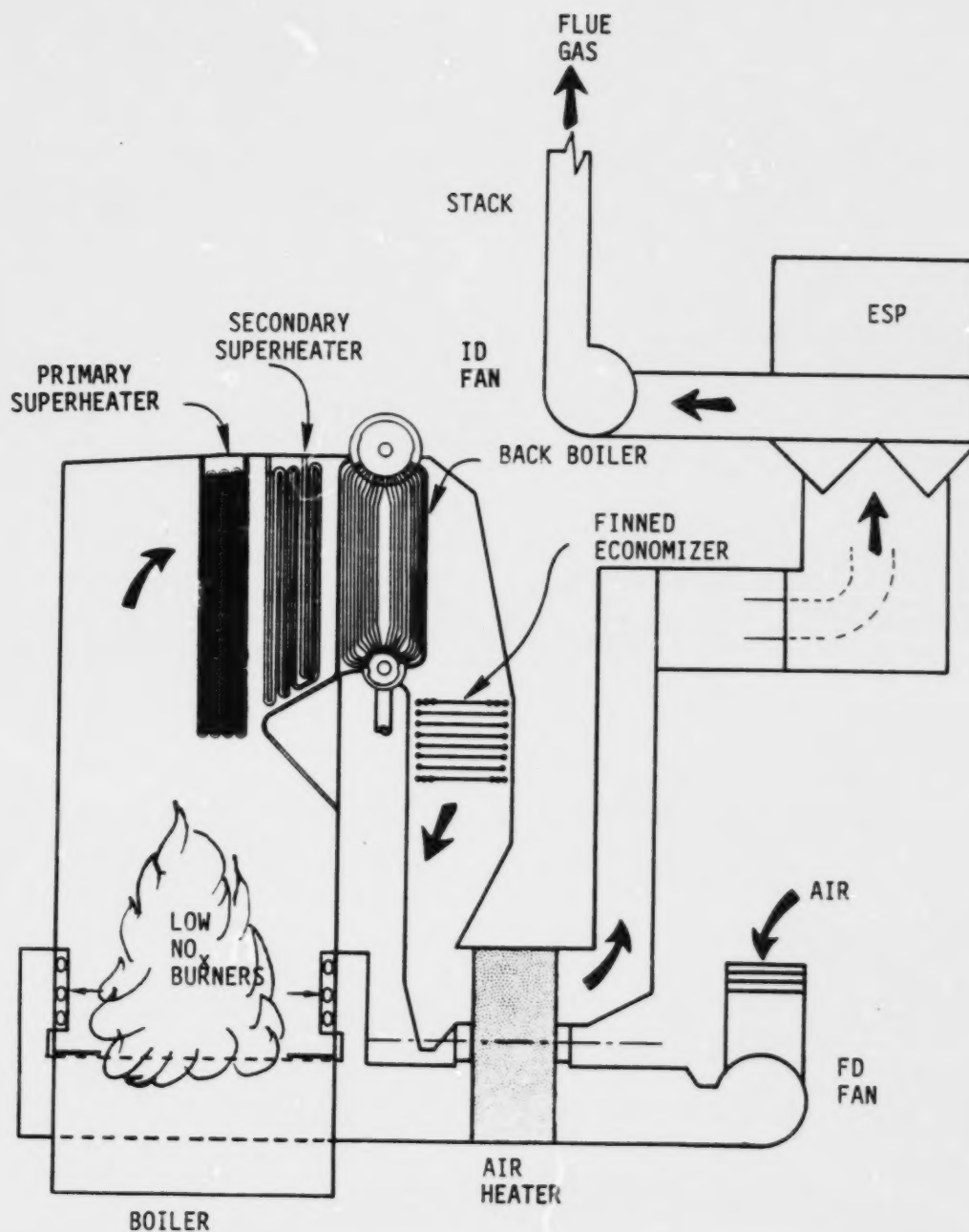


Figure 1. Richmond Power and Light Whitewater Valley Unit 2.



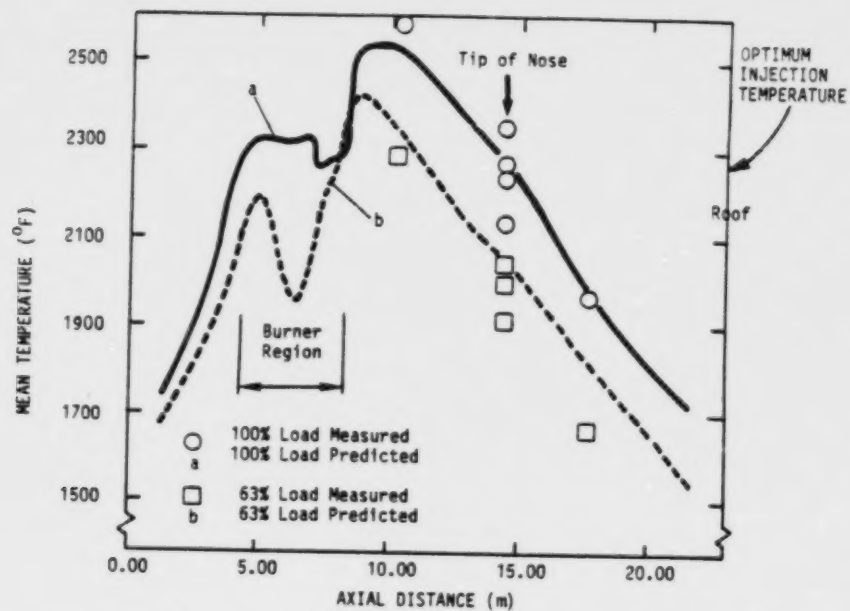


Figure 2. Comparison of mean temperatures ( $^{\circ}\text{F}$ ) predicted for 100% and 63% loads with measured data points.

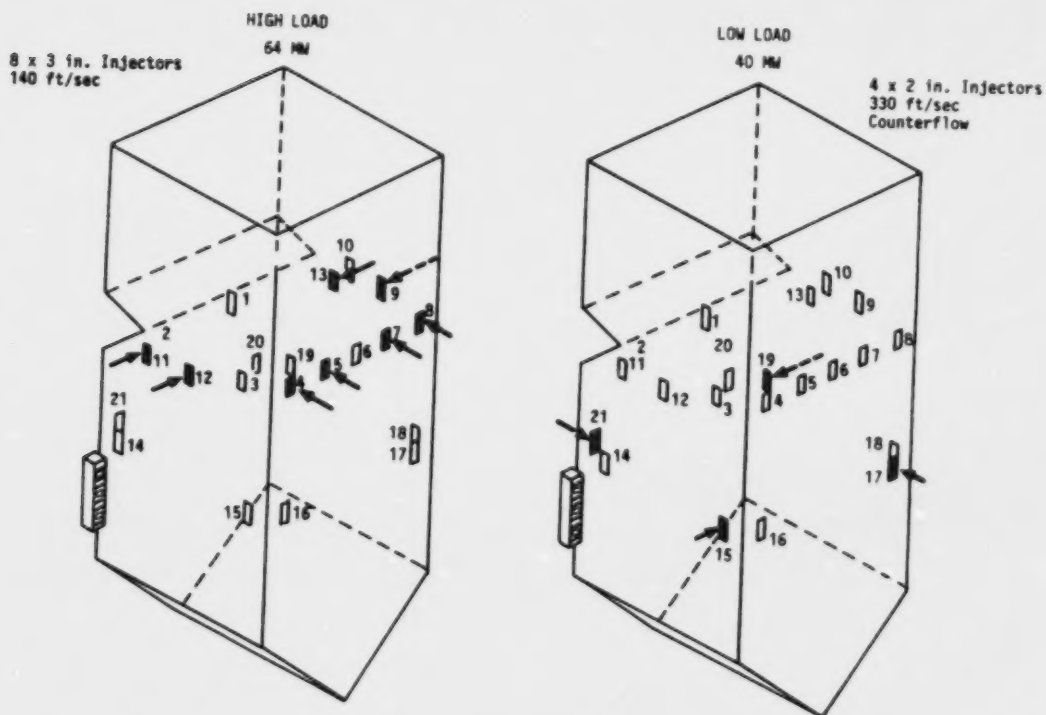


Figure 3. Sorbent injector locations.

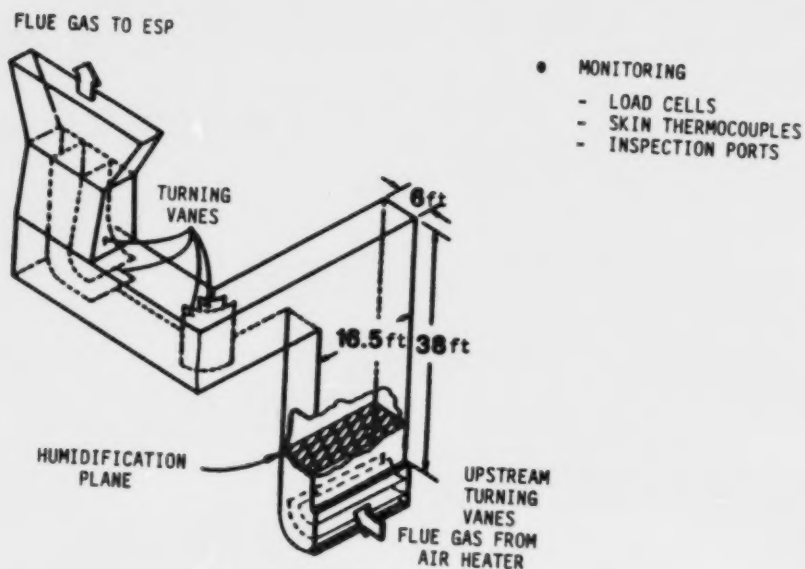


Figure 4. Humidification duct arrangement.

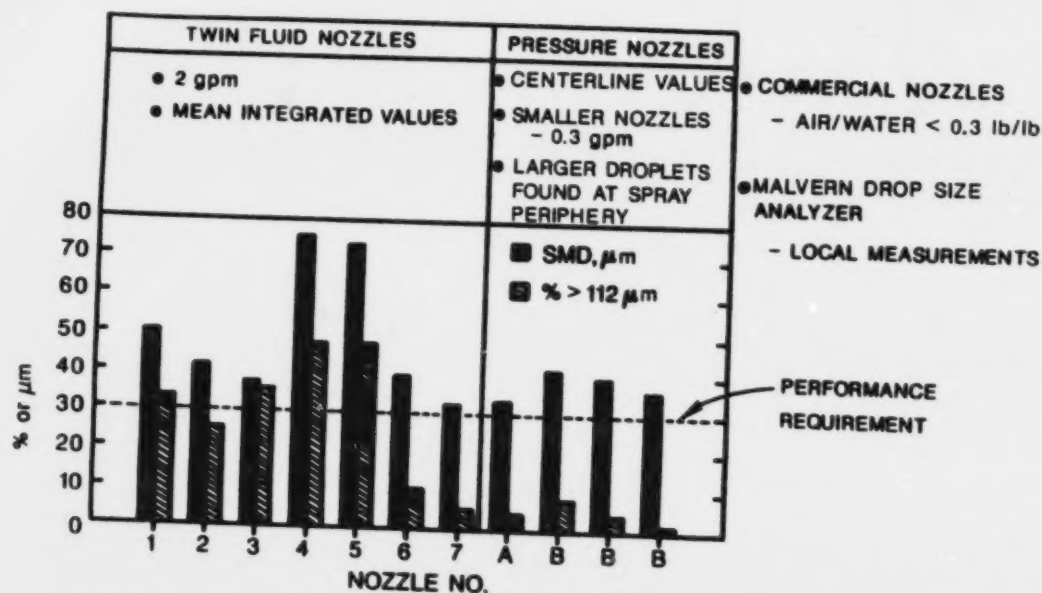


Figure 5. Humidification atomizer screening tests.

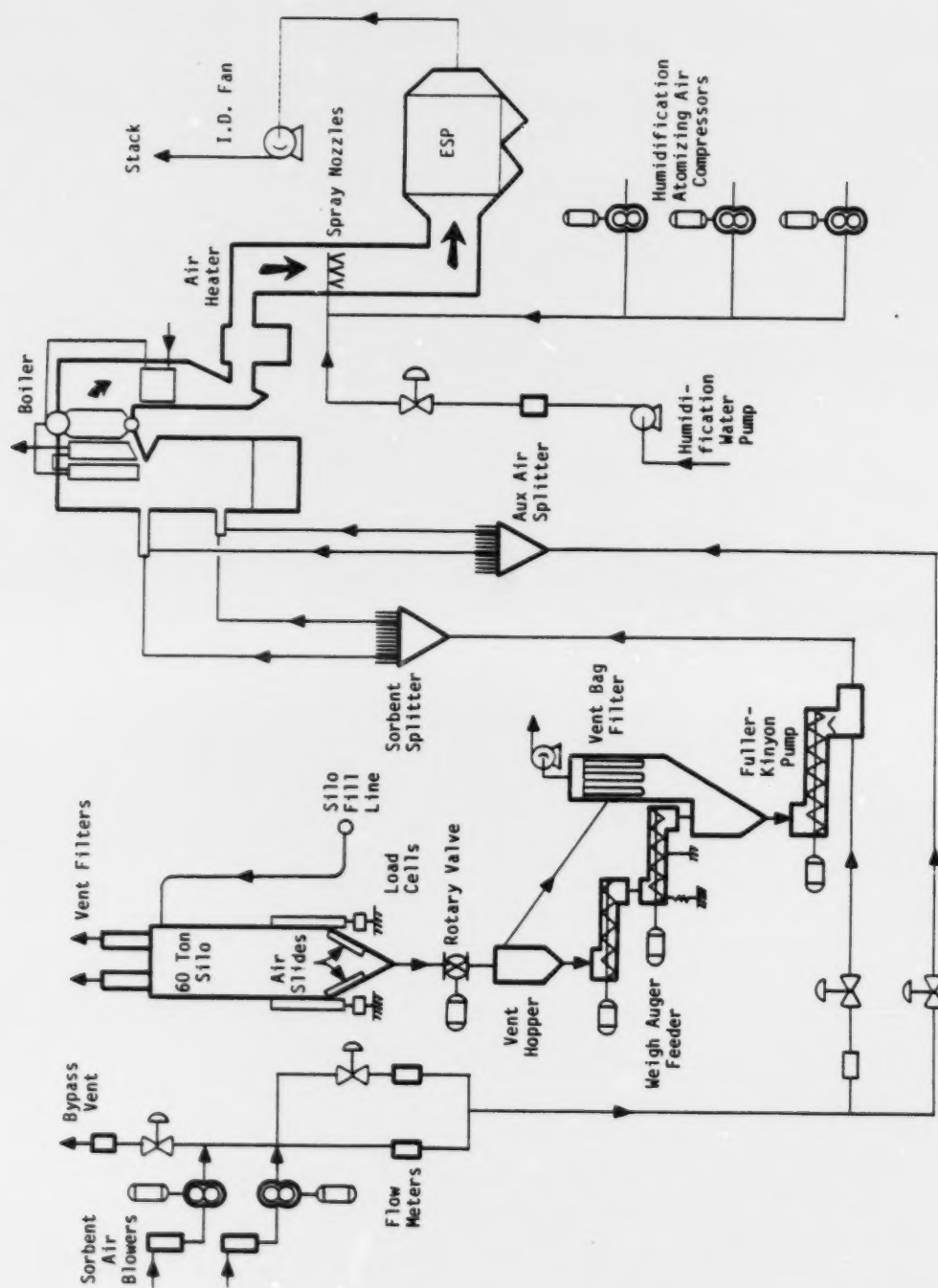


Figure 6. Process flow diagram for Richmond Power and Light LIMB prototype.

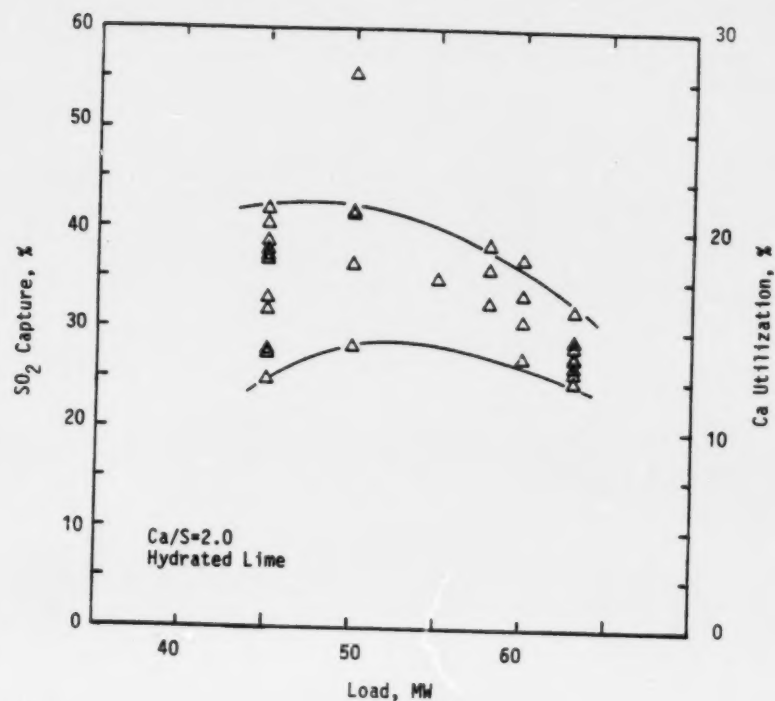


Figure 7. Preliminary SO<sub>2</sub> capture results (upper furnace injection, Marblehead hydrated lime).

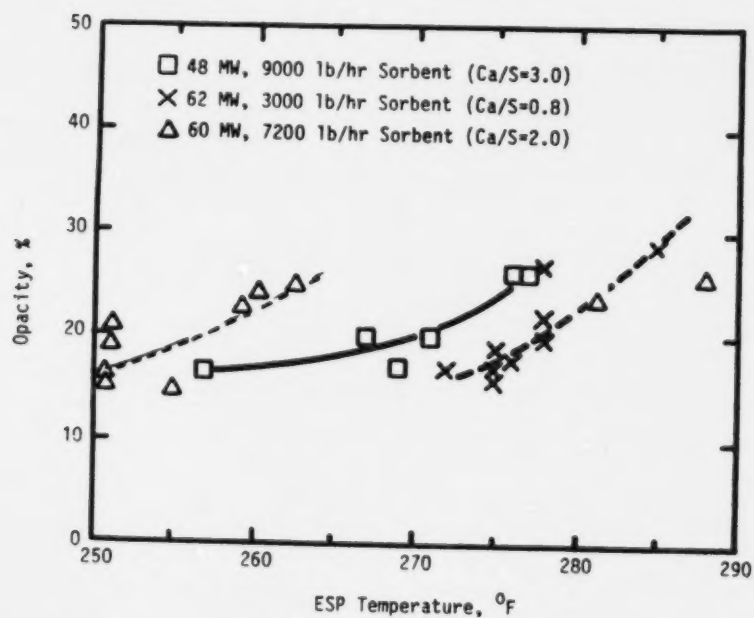


Figure 8. Short term humidification performance.



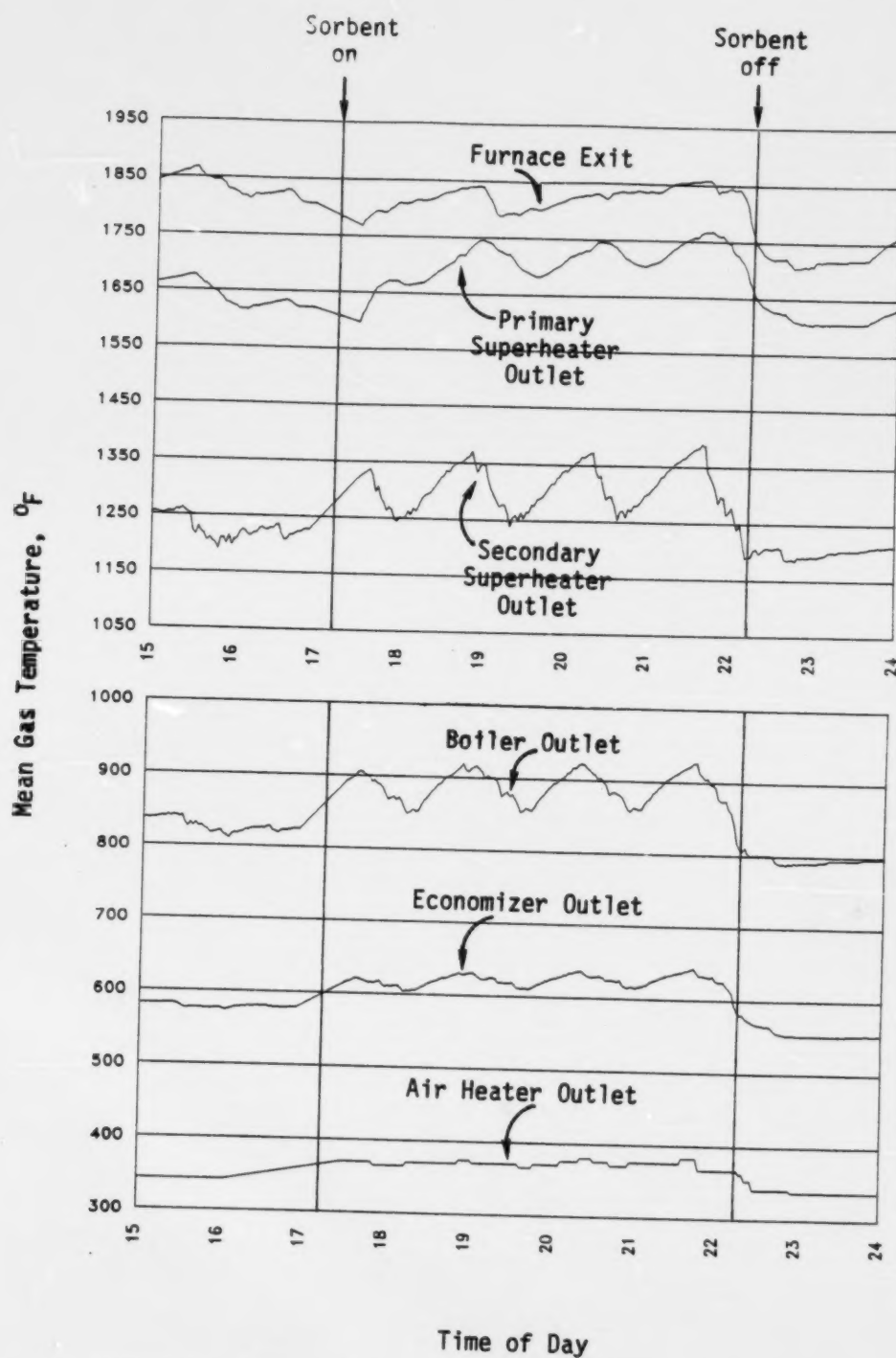


Figure 9. Effect of sorbent injection on boiler gas temperatures (60 MW, Ca/S=2.0).

TABLE 1. DESIGN CHARACTERISTICS

## (a) Boiler Design

| Parameters               | Units           | Design Value      |
|--------------------------|-----------------|-------------------|
| Maximum Capacity Rating  | MW              | 61                |
| Steam Flow               | lb/hr           | 540,000           |
| Heat Input               | Btu/hr          | $651 \times 10^6$ |
| Steam Pressure           | psig            | 1326              |
| Steam Temperature        | °F              | 955               |
| Heating Surface - Boiler | ft <sup>2</sup> | 16,285            |
| - Waterwall              | ft <sup>2</sup> | 9,223             |
| - Economizer             | ft <sup>2</sup> | 20,400            |
| Furnace Width            | ft              | 24.7              |
| Furnace Depth            | ft              | 26.9              |

## (b) ESP Design Characteristics

| Parameters                     | Units                      | Design Value   |
|--------------------------------|----------------------------|----------------|
| Gas Volume                     | acfm                       | 227,000        |
| Gas Temperature                | °F                         | 285            |
| Collection Area (total)        | ft <sup>2</sup>            | 45,000         |
| Specific Collection Area (SCA) | ft <sup>2</sup> /1000 acfm | 198            |
| Inlet Fly Ash Flow             | lb/hr                      | 5,000          |
| Inlet Fly Ash Loading          | gr/ft <sup>3</sup>         | 3.9            |
| Gas Velocity                   | ft/sec                     | 5.5            |
| Residence Time                 | sec                        | 5.46           |
| Number of Fields (in series)   |                            | 4              |
| Plate Length (each field)      | ft                         | 7.5            |
| Plate Height                   | ft                         | 30             |
| Overall Aspect Ratio           |                            | 1.0            |
| Number of Plates (each field)  |                            | 25             |
| Plate - Plate Spacing          | in.                        | 11             |
| Corona Wire Type               |                            | Twisted Square |
| Support                        |                            | Mast           |
| Mast Spacing                   | in.                        | 14-3/8         |
| Number of Wires per Mast       |                            | 2              |
| Wire - Mast Spacing            | in.                        | 3-1/2          |

TABLE 2. SUMMARY OF FEED SYSTEM MODIFICATIONS

|                                                                                       | Original                                                                                       | Modified                                                                                                                                 |
|---------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------|
| Storage Silo<br>Operation<br>Deliveries - Min<br>- Norm                               | Continuous<br>≈ 8 hr<br>≈ 24 hr                                                                | Continuous<br>≈ 8 hr<br>≈ 24 hr                                                                                                          |
| Silo Discharge<br>Hopper<br>Flow Aids<br>Control                                      | 60° w/Air Slides<br>45° Vibrating Discharger<br>Slide Gate                                     | 60° w/Air Slides<br>60° Cone w/Air Slides<br>Rotary Valve                                                                                |
| Feeder - Type<br>- Accuracy<br>- Feedrate                                             | Weigh Auger<br>± 2%<br>2000 - 30,000 lb/hr                                                     | Weigh Auger<br>± 5%<br>2000 - 23,000 lb/hr                                                                                               |
| Transport Air Supply                                                                  | 12 psig Positive Displace-<br>ment Blower                                                      | 12 psig PD Blowers                                                                                                                       |
| Line Injection<br>Air Leakage<br>Transport Lines<br>Solids/Air Ratio<br>Injection Air | Rotary Valve<br>Small Vent to Silo<br>5", 8" Spiral w/Short Elbows<br>1:1<br>Same as Transport | Fuller Kinyon Pump<br>Large Vent to Baghouse<br>5" Rubber Hose<br>0.5 - 6.0<br>12 psig Positive Dis-<br>placement Blower<br>Modified EER |
| Splitters                                                                             | EER                                                                                            |                                                                                                                                          |
| Injectors                                                                             | Round Tubing                                                                                   | Round Tubing with Con-<br>centric transport/<br>Aux Air                                                                                  |
| Injection Velocity                                                                    | 100-300 ft/sec                                                                                 | 100-300 ft/sec                                                                                                                           |

TABLE 3. SORBENT CHARACTERISTICS

| Parameter                             | Units              | USG<br>Limestone | Marblehead (MI)<br>Calcitic<br>Hydrate | Linwood<br>Calcitic<br>Hydrate | Corson<br>(Type N)<br>Dolomitic<br>Hydrate | Marblehead (MI)<br>Calcitic Hydrate<br>(Promoted) |
|---------------------------------------|--------------------|------------------|----------------------------------------|--------------------------------|--------------------------------------------|---------------------------------------------------|
| Surface Area                          | m <sup>2</sup> /g  | 0.87             | 15.54                                  | 16.0                           | 9.75                                       | 11.4                                              |
| Density                               | g/cm <sup>3</sup>  | (2.7)**          | 2.26                                   | 2.30                           | (2.5)                                      | --                                                |
| Median Particle Size                  | μm                 | (10)             | 4.1                                    | 2.2                            | (6)                                        | --                                                |
| Elemental Analysis                    |                    |                  |                                        |                                |                                            |                                                   |
| Ca                                    | wt %               | 38.5             | 50.7                                   | 48.7                           | 33.5                                       | 51.3                                              |
| Mg                                    |                    | (0.53)           | 0.64                                   | 0.97                           | (20.0)                                     | (0.6)                                             |
| Na                                    |                    | --               | 0.01                                   | 0.01                           | --                                         | (0.01)                                            |
| Si                                    |                    | --               | 0.47                                   | 0.66                           | (0.7)                                      | (0.5)                                             |
| Al                                    |                    | (0.16)           | 0.15                                   | 0.18                           | --                                         | (0.1)                                             |
| Fe                                    |                    | (0.07)           | 0.38                                   | 0.27                           | --                                         | (0.4)                                             |
| C                                     |                    | 11.89            | 0.69                                   | 0.33                           | 0.56                                       | 0.99                                              |
| H                                     |                    | 0.11             | 2.32                                   | 2.48                           | 1.39                                       | 2.29                                              |
| N                                     |                    | --               | 0.07                                   | 0.04                           | 0.01                                       | 0.02                                              |
| S                                     | 0.02               | 0.06             | --                                     | --                             | (0.06)                                     |                                                   |
| Bulk Density                          |                    |                  |                                        |                                |                                            |                                                   |
| - Loose                               | lb/ft <sup>3</sup> | 55-70            | 23                                     | --                             | 29                                         | --                                                |
| - Settled                             | lb/ft <sup>3</sup> | 95               | 33                                     | 30-33                          | 56                                         | --                                                |
| SO <sub>2</sub> Capture<br>(Ca/S = 2) |                    |                  |                                        |                                |                                            |                                                   |
| - Gas                                 | %                  | 27               | 51                                     | 58                             | 72                                         | --                                                |
| - Coal                                | %                  | 33               | 50                                     | 57                             | 72                                         | --                                                |

\* Bench Scale

\*\* ( ) Estimated



TABLE 4. PREDICTED SO<sub>2</sub> REMOVAL (Ca/S = 2.0)

| Load<br>Injection Point | 100%<br>Nose | 100%<br>10 m | 63%<br>Nose | 63%<br>10 m |
|-------------------------|--------------|--------------|-------------|-------------|
| APG Limestone           | 33*          | 25           | 37          | 31          |
| Linwood Hydrate         | 54           | 45           | 50          | 63          |
| Marblehead Hydrate      | 45           | 39           | 43          | 51          |
| Corson Dolomitic        | 65           | 54           | 62          | 75          |

\* Percent removal

DEVELOPMENT AND DEMONSTRATION OF SORBENT INJECTION  
FOR SO<sub>2</sub> CONTROL ON TANGENTIALLY COAL-FIRED BOILERS

M. R. Gogineni, J. P. Clark, J. L. Marion, R. W. Koucky,  
D. K. Anderson, A. F. Kwasnik  
Combustion Engineering, Inc.  
Windsor, Connecticut 06095

E. Gootzait  
Virginia Power Company  
Yorktown, Virginia 23690

D. G. Lachapelle, S. L. Rakes  
U.S. Environmental Protection Agency  
Air and Energy Engineering Research Laboratory  
Research Triangle Park, North Carolina 27711

ABSTRACT

Combustion Engineering, under EPA sponsorship, has initiated a program to demonstrate furnace sorbent injection on a tangentially fired, coal-burning utility boiler, Virginia Power's 180 MW(e) Yorktown Unit No. 2. The overall objective of the program is to demonstrate significant reductions in SO<sub>2</sub> and NO<sub>x</sub> while minimizing any negative impacts on boiler performance.

The demonstration program includes unit-specific aerodynamic flow modeling and pilot-scale combustion testing to identify and verify optimum sorbent injection and NO<sub>x</sub> reduction approaches for Yorktown. The procedures which are being used to select and optimize the sorbent injection system for Yorktown were established under a laboratory program recently completed by Combustion Engineering, also under EPA sponsorship, to develop sorbent injection criteria for controlling SO<sub>2</sub> from tangentially fired coal boilers. The program will culminate with an 8-month demonstration test at Yorktown. Flue gas humidification and low temperature simulated sorbent recycle for enhanced SO<sub>2</sub> removal will also be demonstrated during the 8-month test.

The overall demonstration program organization and schedule, plus the results of laboratory testing under both the demonstration and sorbent injection criteria development programs, are discussed in the paper.

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.

## INTRODUCTION

Furnace sorbent injection is under development as an approach for  $\text{SO}_2$  control for coal-burning utility boilers. The process is attractive for retrofit of existing boilers since the capital equipment requirements and overall sulfur reduction costs per ton of sulfur removed are less than for other options such as flue gas desulfurization<sup>(1)</sup>. In the furnace sorbent injection process, sorbent, usually calcium hydroxide  $[\text{Ca}(\text{OH})_2]$  or limestone ( $\text{CaCO}_3$ ), is injected above the flame zone and mixes with the combustion products containing sulfur dioxide ( $\text{SO}_2$ ). The  $\text{SO}_2$  reacts chemically with lime ( $\text{CaO}$ ), formed from the sorbent, to make solid calcium sulfate ( $\text{CaSO}_4$ ). The  $\text{CaSO}_4$  is removed with flyash in particulate collection equipment.

A significant fraction of the existing boilers in the United States that would be candidates for retrofit with sorbent injection technology are tangentially fired units. Tangential firing introduces special considerations for the design of sorbent injection systems due to the unique aerodynamics in the sorbent mixing region of tangentially fired furnaces. The U.S. Environmental Protection Agency (EPA) recognized this fact when it awarded Combustion Engineering (C-E) a contract for Demonstration of Sorbent Injection Technology on a Tangentially Coal-Fired Utility Boiler (EPA Contract 68-02-4275) on June 12, 1987. This demonstration program, which complements an ongoing sorbent injection demonstration program for wall-fired boilers at Ohio Edison's Edgewater Station, follows the recently completed program for Development of Sorbent Injection Criteria for Sulfur Oxides Control from Tangentially Fired Coal Boilers performed by Combustion Engineering under EPA sponsorship (EPA Contract 68-02-4224). This paper presents an overview of the tangentially fired sorbent injection demonstration program, as well as the results of pilot-scale combustion testing under the earlier Sorbent Injection Criteria Development program.

## DISCUSSION

C-E is conducting a program to demonstrate furnace sorbent injection on a tangentially fired, coal-burning utility boiler. The overall objective of the program is to demonstrate significant reduction (approximately 50%) in  $\text{SO}_2$  and  $\text{NO}_x$  while minimizing any negative impacts on boiler performance during long-term operation of an integrated sorbent injection system and a low- $\text{NO}_x$  firing system. The program will address technical sorbent injection-related problem areas, such as sorbent handling and injection, precipitator performance, ash handling and disposal, and the possibility of convective pass fouling over a period of operation sufficient to establish cost-effective resolutions.

The project team consists of C-E (program management, process equipment design/installation/operation), Virginia Power (host utility), Stone & Webster and Lummus Crest (balance-of-plant engineering), and Radian Corporation (emission testing). The project team members are cofunding the project with the EPA.

## HOST UNIT DESCRIPTION

The demonstration program will be conducted at Virginia Power's Yorktown Unit No. 2. An aerial view of the Yorktown Power Station, located in Yorktown, Virginia, is shown in Figure 1. Unit No. 2 is to the right of the smaller stack.

Yorktown Unit No. 2 is a tangentially fired, Controlled Circulation<sup>®</sup> reheat boiler with a current rating of 180 MWe. A side elevation of Unit No. 2 is shown in Figure 2. Pertinent data describing Unit No. 2 are presented in Table

1. Unit No. 2 features a divided furnace with a water-cooled centerwall, with four levels of tilting tangential burners on each side of the dividing wall.

Yorktown Unit No. 2 is equipped with two Ljungstrom® regenerative air heaters. Flue gas from these air heaters enters the electrostatic precipitator (ESP) via separate inlet ducts. The ESP, with a specific collection area (SCA, square feet of collecting area per thousand actual cubic feet per minute of gas treated) of 720, was installed in 1985. When combined with humidification of the flue gas, this large capacity should permit operational flexibility during the demonstration test. Material collected in the ESP hoppers is pneumatically transported to an ash silo, from which it is trucked to a Virginia Power-owned landfill disposal site 2 miles away.

#### PROGRAM DESCRIPTION

The demonstration program is divided into six tasks:

- Task 1 Program Management
- Task 2 Development of Preliminary LIMB (Limestone Injection Multistage Burner) Concept
- Task 3 Determination of Baseline Conditions
- Task 4 Conduct of Demonstration Program
- Task 5 Preparation of Recommendations and Guidelines for LIMB Commercialization
- Task 6 Site Restoration

The Program Schedule is shown in Figure 3. Work on the contract started in June 1987 and is scheduled to be completed in November 1992. Baseline testing without sorbent injection is scheduled for late 1989. Demonstration testing is scheduled for 1991. A medium sulfur (2.0 to 2.5% sulfur) eastern bituminous coal will be used. Atmospheric hydrated lime  $[\text{Ca}(\text{OH})_2]$  will be used as the sorbent.

Each of the tasks is briefly discussed below. The results of pilot-scale testing under the Sorbent Injection Design Criteria contract are also discussed.

#### PROGRAM MANAGEMENT

A host site agreement between C-E and Virginia Power is in place. Several activities are under way under this agreement, including identification of candidate coal and sorbent suppliers and application for required permits and variances.

This tangentially fired demonstration program is, sequentially, the third full-scale EPA-sponsored sorbent injection program, following the wall-fired demonstration program at Ohio Edison's Edgewater Station and the tangentially fired prototype test program at Richmond (Indiana) Power and Light (RP&L). Information obtained during these programs, such as ESP performance enhancements, sorbent handling requirements, and waste handling and disposal considerations, will be incorporated into the Yorktown demonstration program.

#### DEVELOPMENT OF PRELIMINARY LIMB CONCEPT

The preliminary approach for the sorbent injection system, low-NO<sub>x</sub> firing equipment, and flue gas humidification system are being established under this task. In support of this effort, site-specific studies are being conducted,



both on-site at Yorktown and off-site at C-E's Kreisinger Development Laboratory (KDL) in Windsor, Connecticut.

#### On-Site Studies

Extensive temperature traversing was conducted on Yorktown Unit No. 2 at both maximum continuous rating (MCR) and 70% MCR conditions to define temperature profiles and identify the critical temperature zone for sorbent injection. Figure 4 shows the critical SO<sub>2</sub> capture window (2300 to 1650°F) at MCR and 70% MCR. This temperature information was used to establish candidate injection locations for cold flow modeling and pilot-scale combustion testing at KDL.

Gas composition (NO<sub>x</sub>, O<sub>2</sub>, CO) was monitored during the tests. Coal and ash samples were obtained and analyzed at KDL. This information was obtained to support the specification of a low-NO<sub>x</sub> firing system for Yorktown.

#### Off-Site Cold Flow Modeling Studies

Various candidate sorbent injection configurations were evaluated for penetration, dispersion, and mixing using isothermal flow modeling to identify optimum locations and methods for demonstration testing at Virginia Power's Yorktown Unit No. 2. A 1/9-scale model of Yorktown Unit No. 2 was constructed (Figure 5). The entire right-hand cell of Unit No. 2 was modeled from hopper bottom to upper furnace heat transfer surfaces, including the demonstration low-NO<sub>x</sub> firing system. Only the right-hand cell was modeled, since, in an "eight<sup>x</sup> corner" divided furnace, the left-hand and right-hand furnace cells are mirror images of one another.

A large number of candidate sorbent injection configurations were evaluated in the flow model. Figure 6 summarizes the sorbent injection locations investigated. Specific injection configurations involved combinations of injection locations utilizing four, six, and eight simultaneous injectors per furnace cell. Injector locations were selected at those elevations in the boiler where furnace gas temperatures ranged from 2200 to 2400°F at either 70% or 100% MCR operating conditions. Previous programs<sup>(2,3)</sup> have identified these temperatures as optimum for calcium hydrate injection. Rapid mixing of sorbent and furnace gases at the injection elevation is also critical in order to maximize SO<sub>2</sub> capture within the temperature window of 2300 to 1650°F where the sulfation reaction proceeds sufficiently rapidly to have significance in suspension-fired boilers.

Injection configurations tested in the Yorktown boiler model were based on experience gained by C-E under earlier sorbent injection programs, including the program to develop sorbent injection design criteria for tangentially fired coal boilers (EPA Contract 68-02-4224)<sup>(4)</sup>. A brief discussion of this previous program is provided later in this paper. In developing the matrix of tested injection configurations, the particular flow patterns of Yorktown Unit No. 2 were also carefully considered. Figure 7 shows the tangential velocity vectors at a cross-sectional plane in the Yorktown furnace model located between the fuel admission assemblies and the furnace arch. The swirling flow field is evident. By comparison to Figure 8, a similar cross section located in a non-divided "four corner" tangentially fired boiler, the oval fireball pattern of Yorktown Unit No. 2 can be deduced. Such furnace flow patterns are typical of C-E divided furnaces due to the modified placement of the fuel admission assemblies away from the corners and onto the front and rear walls to accommodate the division wall.

Sorbent injection configurations with the best mixing performance utilized several injection locations, with location-specific injection angles for each location, to accommodate the oval fireball pattern. Optimum injection configurations were identified through several screening levels which initially involved flow visualization analysis and, later, for selected tests, quantitative gas tracer studies. Testing was performed at simulated full load and 70% load furnace operating conditions. A typical sorbent injection flow visualization pattern is shown in Figure 9. The best flow visualization tests were retested using a tracer gas. Concentrations of tracer gas were then mapped at various cross sections of the furnace, including the horizontal furnace outlet plane (HFOP) at the furnace arch and the vertical furnace outlet plane (VFOP) above the arch. The variation in tracer gas concentration across these planes was quantified statistically. An example of these results is shown in Figure 10. Using this approach, the most promising configurations were identified and recommended for the Yorktown demonstration.

Following cold flow model studies, pilot-scale combustion testing was performed in C-E's Boiler Simulation Facility (BSF) to further support the demonstration sorbent injection design. To support the pilot-scale combustion program, discussed below, additional cold flow modeling was performed in a model which closely matched the BSF following modification to simulate, within practical limits, Yorktown Unit No. 2. The cold flow model of the BSF is shown in Figure 11. A similar program of flow visualization and tracer studies was conducted. This effort provided a bridge between cold flow Yorktown results and pilot-scale combustion results, thereby permitting better resolution of the final recommendations for sorbent injection at Yorktown Unit No. 2.

#### Off-Site Combustion Testing

C-E's 15 MW(t) ( $50 \times 10^6$  Btu/hr) BSF was modified to reflect the low-NO<sub>x</sub> firing system, gas time/temperature profile, and upper furnace configuration of Yorktown Unit No. 2. This allowed combustion testing to be conducted with operating conditions similar to those of the Yorktown unit. As with the cold flow model, only the right-hand furnace cell was modeled. Figures 12 and 13 show the BSF test furnace.

Tests are being performed utilizing a candidate demonstration coal and sorbent. The coal specification is:

- 2.0% to 2.5% Sulfur
- 12,500 Btu/lb, minimum
- 11% Ash, maximum
- 8% Moisture, maximum
- 45 Hardgrove Grindability Index, minimum

The sorbent utilized for BSF combustion testing is a high-calcium hydrated lime treated with calcium lignosulfonate supplied by Marblehead Lime Co. A small quantity of untreated hydrate will also be tested for comparison purposes.

Initial combustion testing was performed to characterize and optimize combustion performance and NO<sub>x</sub> emissions with the low-NO<sub>x</sub> firing system. Furnace gas temperatures were then mapped and found to match those of Yorktown. The remainder of the testing at the BSF will investigate and optimize sorbent injection locations and methods recommended under cold-flow modeling. At the time of paper preparation, combustion testing was in progress. Some initial tests, however, achieved 50% SO<sub>2</sub> capture at a Ca/S molar ratio of 2:1. These results were obtained by injection configurations that injected into furnace gases ranging in temperature from 2200 to 2400°F. Further sorbent injection tests are in progress.

## Development of Sorbent Injection Criteria Program

This program was performed prior to the Yorktown boiler demonstration program to establish design criteria for sorbent injection in tangentially fired coal-burning furnaces. The design criteria developed under this program (EPA Contract 68-02-4224)<sup>(4)</sup> is being extensively applied to the current demonstration. This program included cold-flow isothermal modeling, mathematical modeling, combustion testing, and design criteria development tasks.

Cold-flow modeling was analogous to that previously discussed for the demonstration program. Under this program, techniques for injector modeling were developed and then applied toward flow visualization and gas tracer screening of sorbent injector configurations. Sorbent injection configurations were also investigated by computational fluid dynamics (CFD). An example of calculated upper furnace flow patterns and sorbent dispersion utilizing a three-dimensional computer model is presented in Figure 14.

Mathematical modeling also applied proprietary C-E thermal predictive codes to calculate furnace gas time/temperature profiles in the BSF combustion facility. Also under mathematical modeling, an SO<sub>2</sub> capture model was applied which utilizes cold flow mixing data. These various models are being applied to the Yorktown demonstration.

BSF combustion testing was performed using an eastern bituminous coal containing 2.7% to 3.2% sulfur. Over 160 tests were performed, including baseline tests, upper furnace temperature characterization tests, and NO<sub>x</sub> measurements, as well as sorbent injection tests over a range of Ca/S molar<sup>x</sup> ratios, sorbent types, injector tilts, sorbent carrier air velocities, and injector locations. Three sorbents were tested: untreated Black River hydrated lime, untreated Marblehead hydrated lime, and Marblehead hydrated lime treated with calcium lignosulfonate. SO<sub>2</sub> capture varied with sorbent type and test conditions.

Up to 50% SO<sub>2</sub> capture was achieved with lignosulfonate-treated hydrate; the other sorbents produced lower SO<sub>2</sub> captures. This is consistent with results achieved by the EPA during bench-scale testing<sup>(5)</sup>.

Figures 15 and 16 show typical SO<sub>2</sub> captures obtained. Figure 15 shows SO<sub>2</sub> capture obtained with four corner located injectors injecting in a tangential manner below the furnace arch (defined as Location D). This figure shows that SO<sub>2</sub> capture dramatically improved as injector tilt was varied from downward injection (-45°) to upward injection (+45°). This trend is contrary to results obtained from cold-flow testing, which showed that downward tilt produced improved mixing. This discrepancy is due to the effects of furnace gas temperatures. Furnace gas temperatures at Location D averaged about 2500°F. The average furnace gas temperature just 2 feet above Location D (at the bottom of the furnace arch) was 2350°F; 5 feet above Location D (at the centerline of the furnace arch or horizontal furnace outlet plane) it was 2200°F. Downward (-45° tilt) or horizontal (0° tilt) injection at Location D likely exposed the sorbent to excessive furnace gas temperatures (certainly greater than 2400°F). Upward injection (+45° tilt) produced sorbent jets which quickly penetrated into the more favorable furnace gas temperatures of 2350 to 2250°F. These data show that, although Location D was actually lower than the ideal location, injector tilt could be used to compensate by changing the effective sorbent injection elevation.

Figure 16 compares the performance of the three sorbents tested. This comparison was made for injection through four front wall injectors located just above the furnace arch. Although this injection configuration did not produce the highest SO<sub>2</sub> captures, the relative SO<sub>2</sub> capture performance of the three sorbents



is representative. The magnitude of the performance difference was lowest for lower furnace injection and highest with upper furnace injection. The calcium lignosulfonate-treated hydrate obtained from Marblehead Lime Co. was consistently highest in SO<sub>2</sub> capture performance. This sorbent is also a candidate for the Yorktown demonstration program.

#### Preliminary Process Design

Preliminary design of the sorbent handling and injection system and the flue gas humidification system is in progress. Various sorbent conveyance and injection alternatives are being evaluated, incorporating operating experience from C-E's BSF, RP&L, Edgewater, Ontario Hydro's Lakeview Station, and Saskatchewan Power's Boundary Dam Station. Design of the flue gas humidification system will also draw heavily from experience gained at RP&L and Edgewater. Computational fluid dynamics modeling and laboratory testing are being used to establish the preferred humidification approach for Yorktown.

#### DETERMINATION OF BASELINE CONDITIONS

Accurate definition of boiler performance without sorbent injection is critical in determining the effectiveness of this process in controlling SO<sub>2</sub>, as well as in establishing the normal operating characteristics at Yorktown Unit No. 2, including fouling, ash handling and disposal requirements, and ESP performance. C-E will characterize the performance of the unit, with the existing tangential firing system, prior to installation of the sorbent injection and low-NO<sub>x</sub> firing systems. A test of 30 days duration on the higher sulfur demonstration coal will be conducted in late 1989. Extensive emissions and boiler performance data will be acquired throughout the continuous 30-day test. These data will establish a baseline level of boiler performance, including SO<sub>2</sub> and NO<sub>x</sub> emissions, against which subsequent sorbent injection demonstration testing can be compared.

#### CONDUCT OF DEMONSTRATION PROGRAM

Under this task, C-E will carry out the field demonstration portion of this program by retrofitting Yorktown Unit No. 2 with an integrated sorbent injection system, including flue gas humidification, plus firing system modifications, and operating this system continuously for 8 months.

Detailed engineering design of the sorbent injection storage, handling, and injection equipment, flue gas humidification and sorbent recycle equipment, low-NO<sub>x</sub> firing equipment, and boiler modifications will be conducted under this task. <sup>x</sup> Equipment installation and erection is scheduled for 1990. Boiler penetrations and other gas path modifications are scheduled for a planned Unit No. 2 outage during the spring of 1990.

Following an equipment start-up and optimization period, the integrated system will be operated continuously for 8 months over the normal boiler duty cycle. Extensive emissions and boiler performance data will be acquired throughout the demonstration test during four 30-day test periods. The current schedule is to test for 4 months with furnace sorbent injection alone, 2 months with furnace sorbent injection plus flue gas humidification, and 2 months with furnace sorbent injection, flue gas humidification, and injection of fresh sorbent into the ESP inlet ducts to simulate recycle.

Recent testing at RP&L and Edgewater has demonstrated that high concentrations



of calcium products in the flue gas can severely compromise ESP performance. Humidification of the flue gas has been shown to be effective in restoring ESP performance to acceptable levels. Thus, it may be necessary to modify the test schedule at Yorktown to include flue gas humidification throughout the demonstration test.

The data obtained during this demonstration test program will be analyzed and compared against baseline data to establish the effects of sorbent injection on emissions, boiler performance, and waste disposal. Incremental costs associated with the process will be identified.

#### PREPARATION OF RECOMMENDATIONS AND GUIDELINES FOR LIMB COMMERCIALIZATION

C-E will evaluate the results of the Yorktown demonstration program, along with the results of other ongoing test programs (prototype testing at Richmond Power and Light, pilot-scale testing at C-E's BSF), and prepare recommendations and guidelines for the further commercialization and use of the sorbent injection process at other tangentially coal-fired boiler sites.

#### SITE RESTORATION

Upon completion of the demonstration test, Yorktown Unit No. 2 and the Yorktown site will be returned to its original condition, less normal wear and tear, subject to discussions between Virginia Power and the EPA.

#### SUMMARY

With the advent of the acid rain issue, utilities and industry are again investigating sorbent injection as a cost-effective alternative to conventional desulfurization systems for achieving moderate levels of SO<sub>2</sub> removal which would be sufficient to meet SO<sub>2</sub> emission requirements for pre-1971 coal-burning units. This paper has described recent C-E activities in the area of furnace sorbent injection for tangentially fired boilers. Successful completion of the Yorktown demonstration program will firmly establish the viability of this process as an alternative to scrubbers for SO<sub>2</sub> control.

#### UNIT CONVERSION TABLE

| <u>To Convert From</u>      | <u>To</u>           | <u>Multiply By</u>     |
|-----------------------------|---------------------|------------------------|
| British thermal units/pound | joules/kilogram     | $2.326 \times 10^3$    |
| degree Fahrenheit           | degree Celsius      | $(T-32)/1.8$           |
| cubic feet/minute           | cubic meters/second | $4.719 \times 10^{-4}$ |
| square feet                 | square meters       | $9.290 \times 10^{-2}$ |
| feet                        | meters              | 0.3048                 |
| inches                      | centimeters         | 2.540                  |
| miles                       | kilometers          | 1.6093                 |
| pounds/hour                 | kilograms/second    | $1.26 \times 10^{-4}$  |
| pounds/square inch          | pascals             | $6.895 \times 10^3$    |
| feet/second                 | meters/second       | 0.3048                 |

# REFERENCES

1. Clark, J. P., Kokkinos, A., Borio, D. C., Koucky, R. W., Sun, C. Y., "Boiler Design Criteria for Dry Sorbent SO<sub>2</sub> Control with Low-NO<sub>x</sub> Burners," EPA-600/7-86-023 (NTIS PB 86-216-736) (1986).
2. Stern, R. D., "EPA's Program for Evaluation and Demonstration of Low Cost Retrofit LIMB Technology," Proceedings Fourth Annual Pittsburgh Coal Conference, p. 232 (1987).
3. Beittel, R., Bortz, S. J., Offen, G. R., and Drehmel, D. C., "Effects of Injection Temperature and Quench Rate on Sorbent Utilization," Proceedings 1986 Joint Symposium on Dry SO<sub>2</sub> and Simultaneous SO<sub>2</sub>/NO Control Technology, Vol. 1, EPA-600/9-86-029a (NTIS PB 87-120-465) (1986).
4. Koucky, R. W., Marion, J. L., Anderson, D. K., "Development of Sorbent Injection Criteria for Sulfur Oxides Control from Tangentially Fired Coal Boilers," EPA-600/7-88-013 (NTIS PB 88-238-357) (1988).
5. Kirchgessner, D. A., and Lorrain, J. M., "Lignosulfonate-Modified Calcium Hydroxide for Sulfur Dioxide Control," I. and E. C. Research, Vol. 26, p. 2397 (1987).

**TABLE 1. VIRGINIA POWER'S YORKTOWN UNIT NO. 2  
UNIT DESCRIPTION**

|                           |                                                                      |
|---------------------------|----------------------------------------------------------------------|
| <b>Type:</b>              | <b>Tangentially fired, Controlled Circulation® , reheat, divided</b> |
| <b>Rating:</b>            | <b>180 MW maximum capacity;<br/>150 MW rated</b>                     |
| <b>Date in Service:</b>   | <b>January 1959</b>                                                  |
| <b>Steam Conditions:</b>  | <b>2000 psi/1000°F/1000°F</b>                                        |
| <b>Main Steam Flow:</b>   | <b>1,200,000 lb/hr</b>                                               |
| <b>Reheat Steam Flow:</b> | <b>1,060,000 lb/hr</b>                                               |

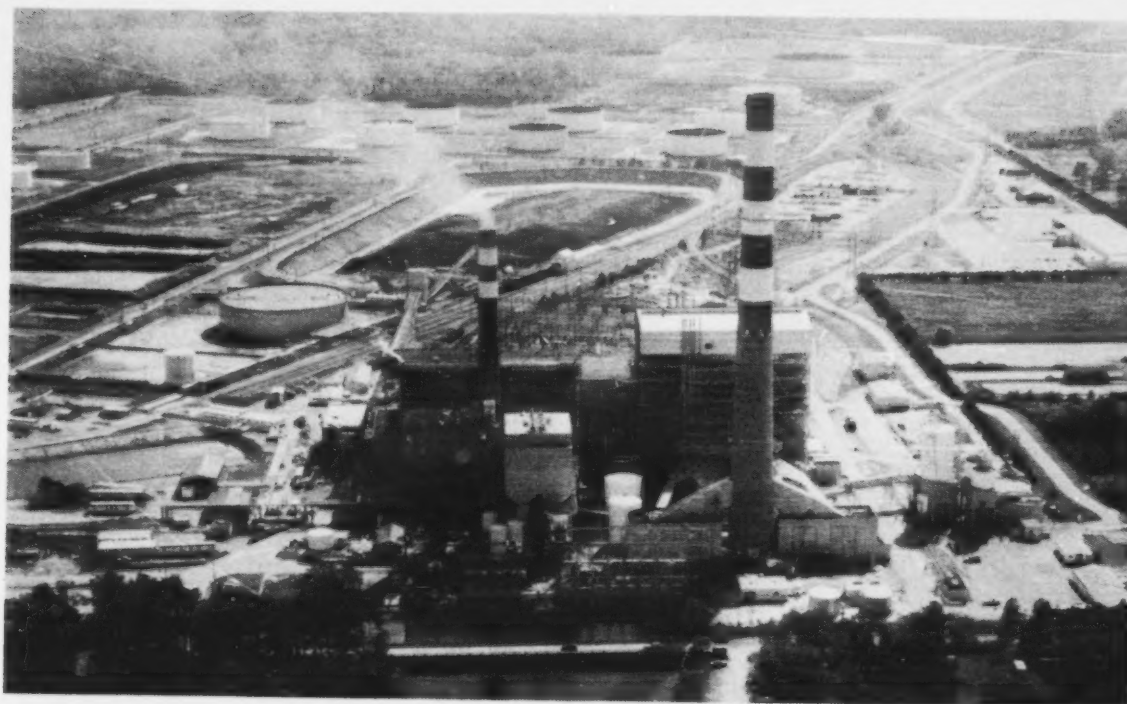


**Figure 1 Virginia Power's Yorktown Power Station**

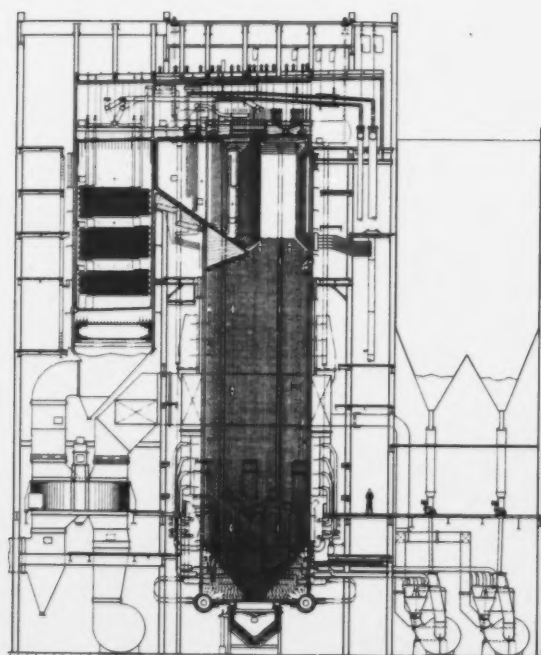


**Figure 2 Side Elevation of Virginia Power's Yorktown Unit No. 2**

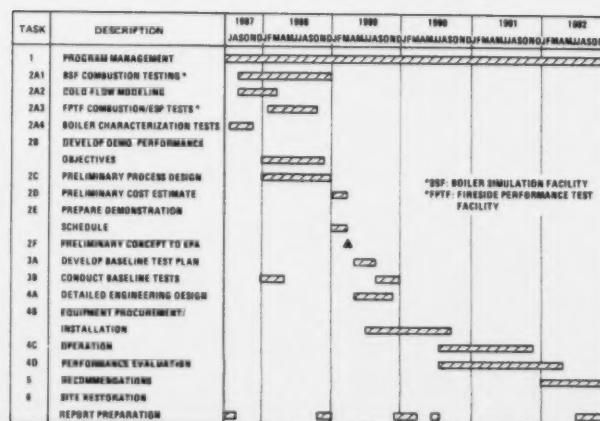


**Figure 3 Program Schedule for EPA Tangentially Fired LIMB Demonstration Program**

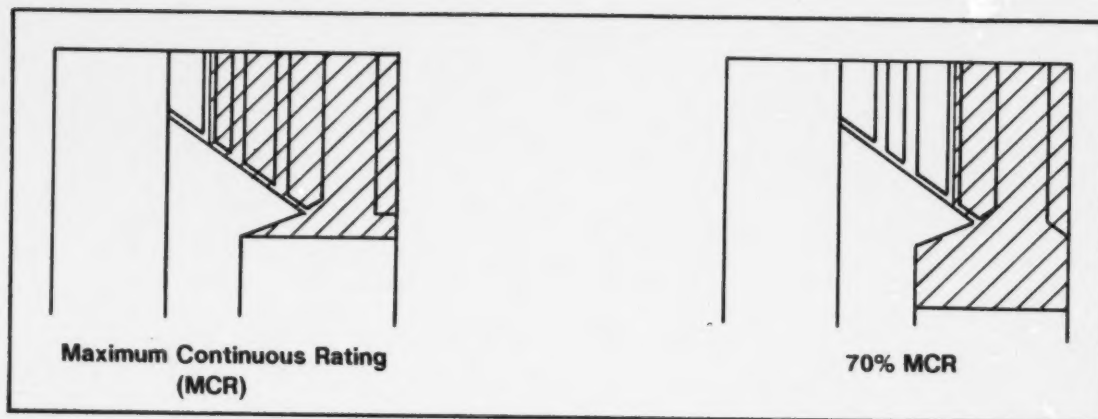


Figure 4 Sulfation Zones for Yorktown Unit No. 2

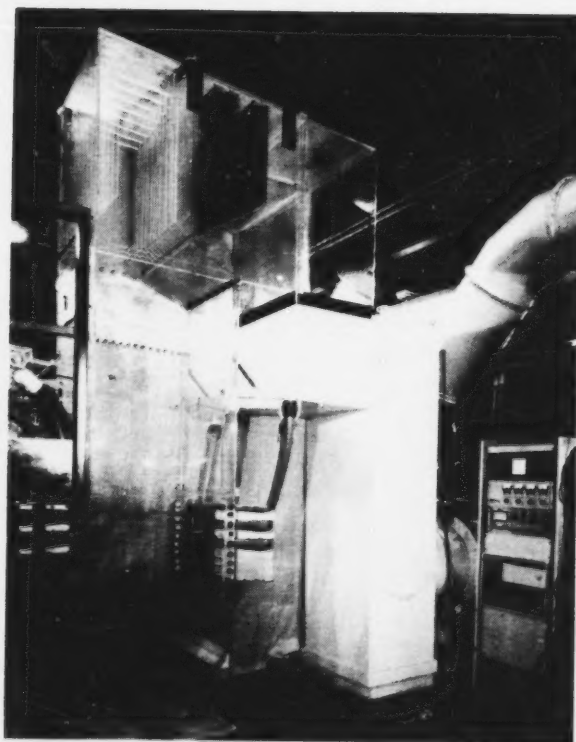


Figure 5 Cold Flow Model of Yorktown Unit No. 2

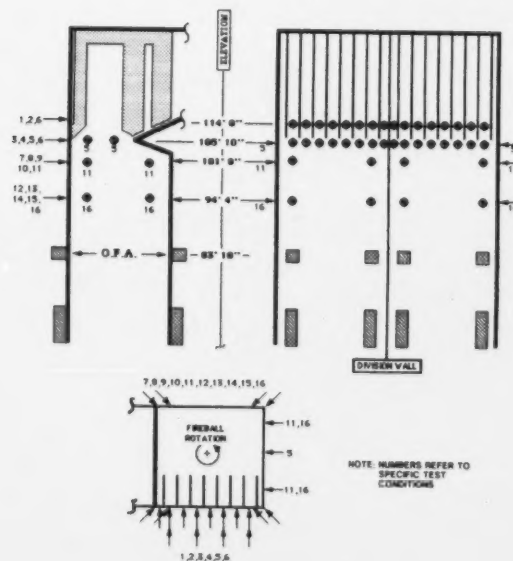


Figure 6 Summary of Investigated Sorbent Injection Location for Yorktown Unit No. 2



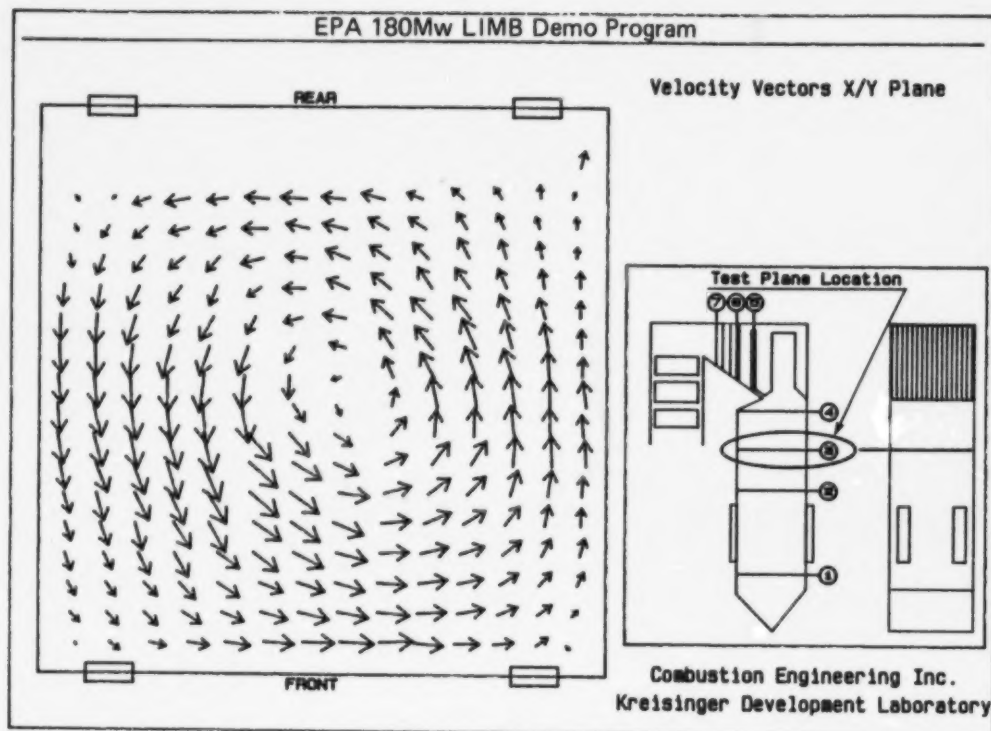


Figure 7 Yorktown Unit No. 2 Flow Model Mid-Furnace Aerodynamic Flow Field

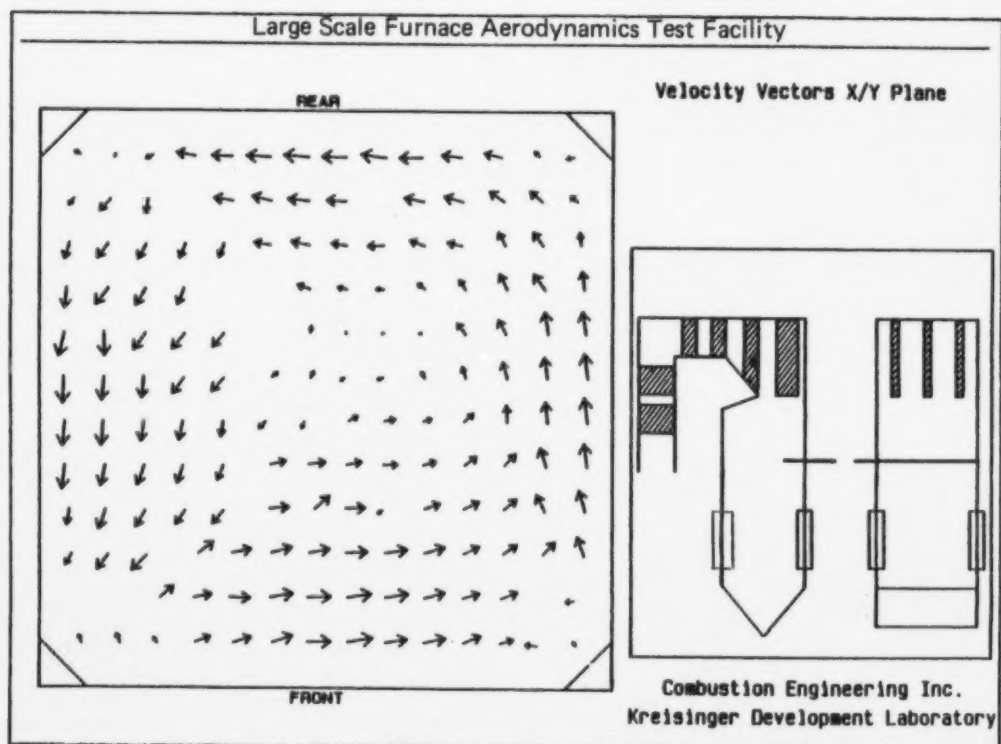


Figure 8 Typical Mid-Furnace Aerodynamic Flow Field

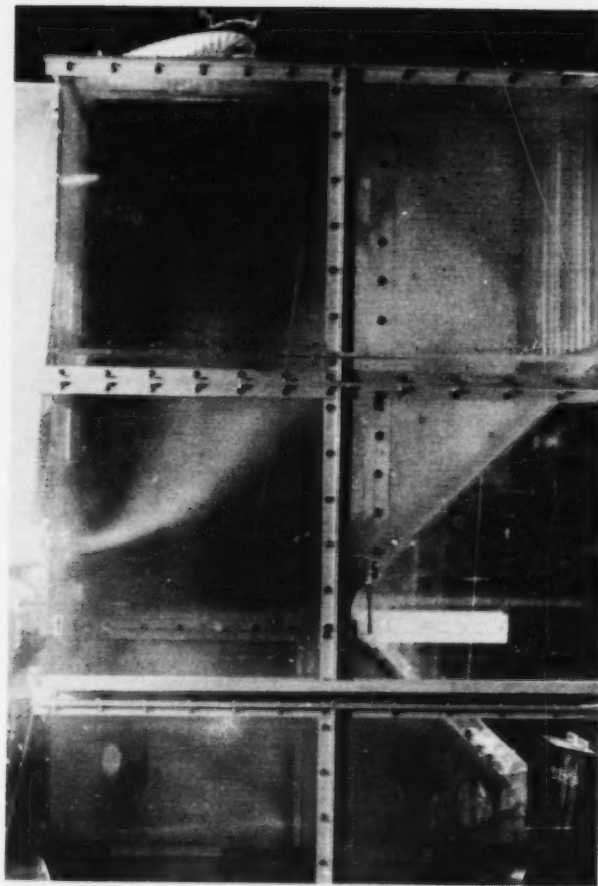


Figure 9 Typical Flow Visualization Test of Front Wall Injection

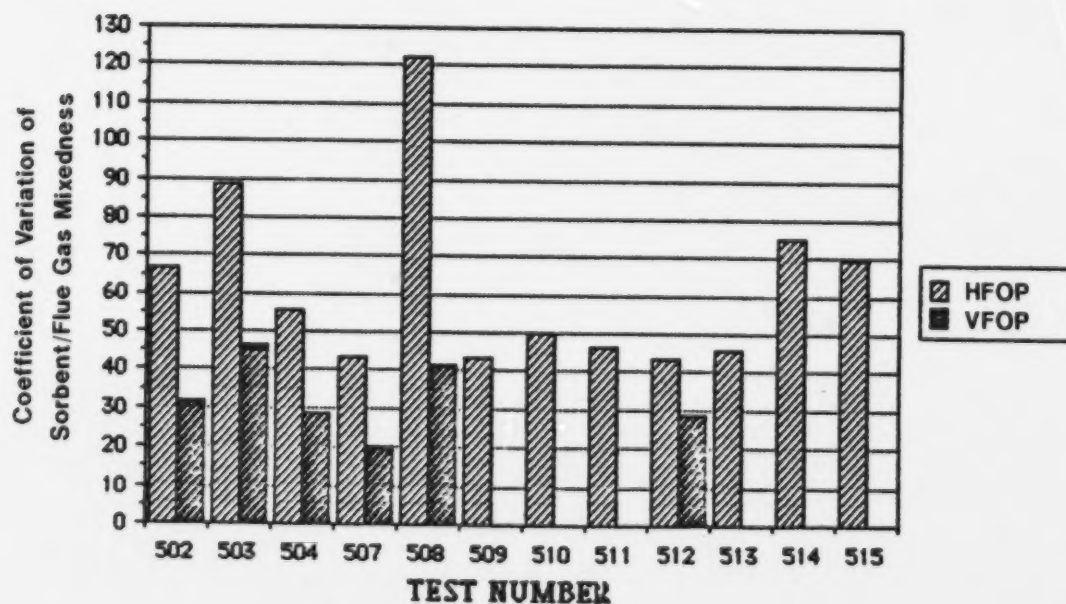


Figure 10 Coefficient of Variation of Sorbent/Flue Gas Mixedness for Yorktown Unit No. 2 Flow Model

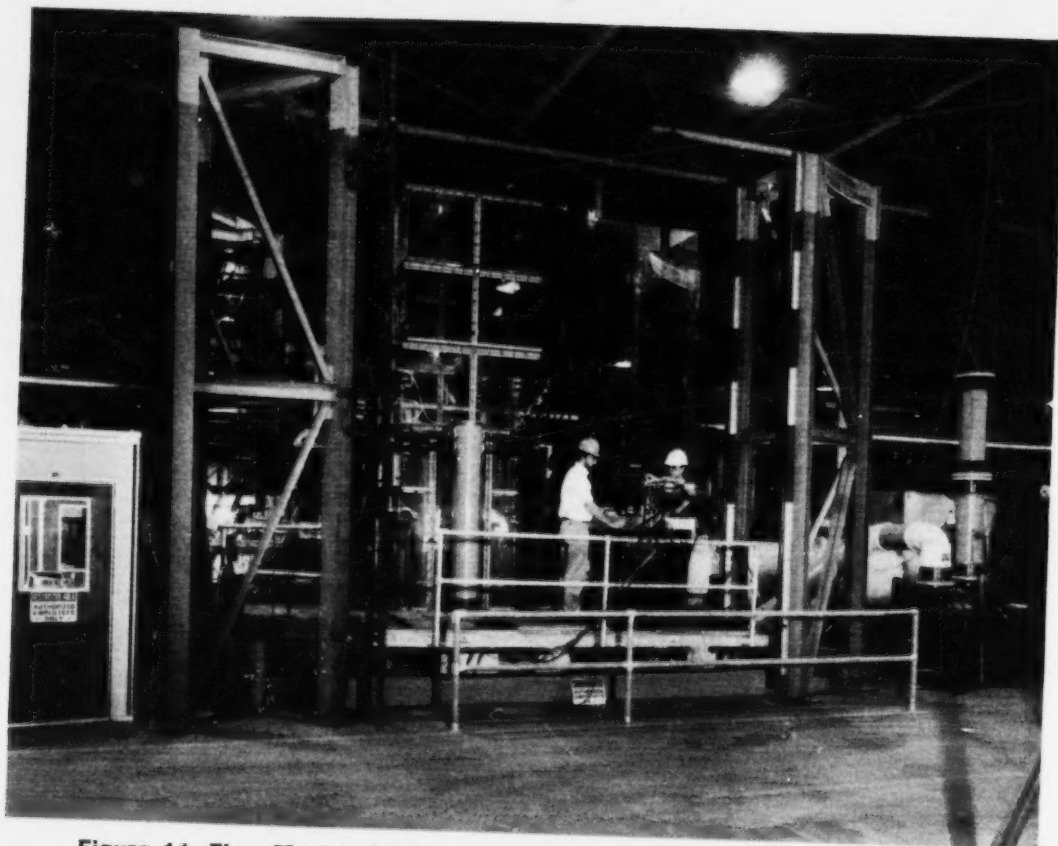


**Figure 11 Flow Model of BSF Configured to Simulate Yorktown Unit No. 2**

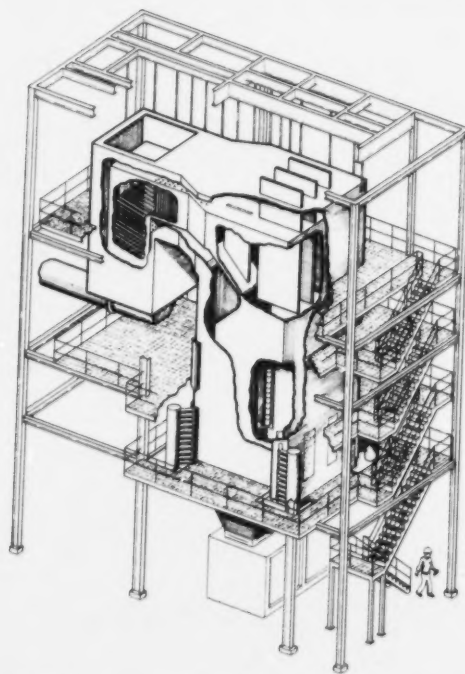


**Figure 12 Combustion Engineering's Boiler Simulation Facility (BSF)**



Figure 13 BSF in Operation

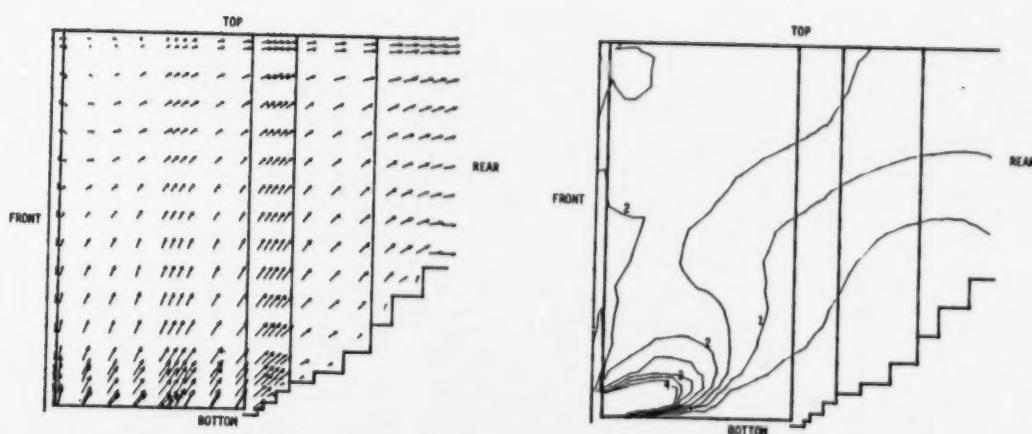


Figure 14 Example of Calculated Flow Patterns and Sorbent Dispersion



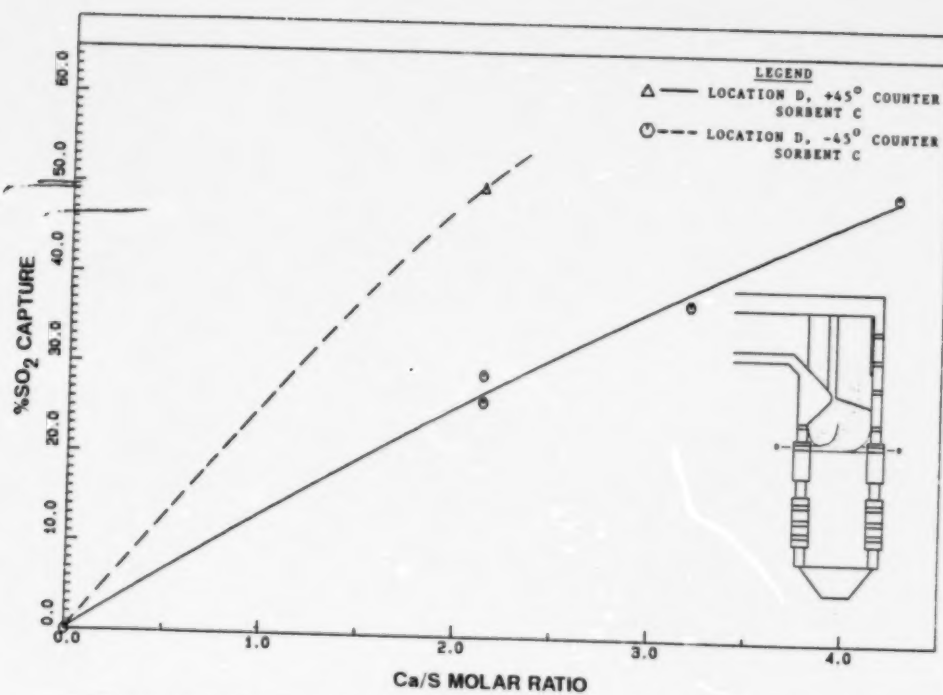


Figure 15 Effect of Injector Tilt on SO<sub>2</sub> Capture During BSF Testing

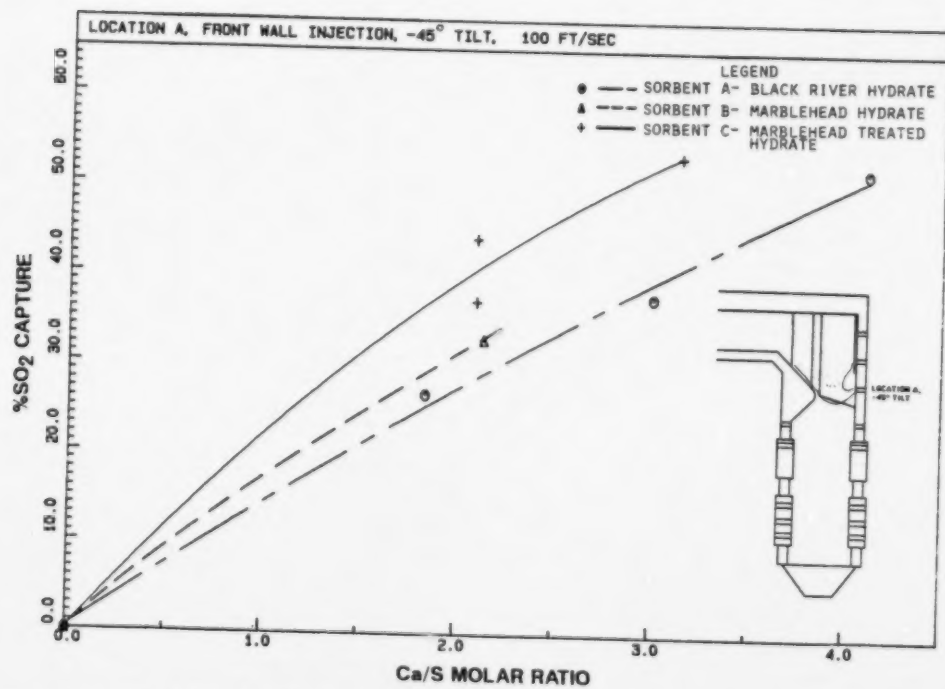


Figure 16 SO<sub>2</sub> Capture Performance Variation with Sorbent Type During BSF Testing

## STATUS OF THE LIFAC SO<sub>2</sub> AND NO<sub>x</sub> REMOVAL PROCESS

by: Timo Kenakkala, M. Sc.  
Tampella Ltd.  
Boiler Division  
Air Pollution Control Department  
Tampere, Finland

Erkki Valimäki, M. Sc. (Eng.)  
Tampella Ltd.  
Boiler Division  
Air Pollution Control Department  
Tampere, Finland

### ABSTRACT

This paper describes briefly the development and current status of the Tampella LIFAC SO<sub>2</sub> removal process, the suitability of the process for different fuels, an indication of NO<sub>x</sub> removal development work employing this system, and the operation and the costs of the process.

The development work of the LIFAC-process started in the early 1980s. After an intensive literature survey and numerous laboratory experiments, a small pilot scale facility was installed on a 250 MW<sub>e</sub> coal-fired boiler. Subsequently, the LIFAC process has been tested both for coal and oil firing in an 8 MW<sub>th</sub> test boiler. The sulfur content of the fuels tested at this facility has varied between 0.5% and 4.3%, including Pittsburgh seam 8 coals from the U.S. The results show that the LIFAC process is suitable and effective in removing over 80% of the SO<sub>2</sub> in flue gas for different fuels irrespective of the sulfur content of the fuel.

The first full-scale LIFAC facility was installed at Imatran Voima Oy's Inkoo power plant in Finland. The first LIFAC system was installed on a portion of a 250 MW<sub>e</sub> coal-fired unit, one of four such units at this plant, and has been in operation since 1985 with a second LIFAC system installed and operating since early 1988. The test results together with the pilot plant tests confirmed the design and operational criteria of the process and mathematical models used in characterizing and designing effective SO<sub>2</sub> removal with this process for different applications. The LIFAC process is now considered fully commercial and is being marketed in Europe and North America for demonstration and operating applications.

## SO<sub>2</sub> EMISSION REDUCTIONS FROM FLUE GASES USING TAMPELLA'S LIFAC PROCESS

### INTRODUCTION

Tampella has been closely involved in the design and commercialization of pollution control equipment to purify the flue gases from industrial and utility facilities since the early 1970s. Since 1972, Tampella has delivered flue gas scrubbers to the pulp and paper industry. In the early 1980s, Tampella's Boiler Division initiated development work that has led to the successful design and commercialization of an efficient, simplified and cost-effective approach to desulfurization of flue gases from power plants. This paper discusses the highlights of this development work and the key features of this new and important process.

The development work directed by Tampella has been carried out intensively in a large number of facilities, by numerous organizations and people testing and developing the desulfurization process in laboratory facilities, pilot scale test boilers, and in full-scale utility power plants. The culmination of this work has been the development of a fully commercialized desulfurization process called Tampella LIFAC<sup>1</sup>. The LIFAC process provides for a much enhanced SO<sub>2</sub> removal when compared to that achieved by other sorbent-injection systems as a result of its unique combination of both in-furnace injection with a subsequent activation reactor for humidification. The end result is a process that achieves 80 to 85% sulfur dioxide removal with commercial grade pulverized limestone at Ca/S molar ratios of 2.0 or less.

The full development of the LIFAC process has required the extensive efforts of not only Tampella Ltd. but various universities and research institutes throughout Finland. The concepts and final process parameters have evolved significantly over time through (1) many intensive laboratory and pilot scale tests, and (2) confirmation and operational refinements developed in a full-scale application of the process at a utility coal-fired power plant.

### BASIS OF THE DEVELOPMENT WORK

The development of the LIFAC desulfurization process (see Figure 1) was initiated in 1984 in Finland by Tampella Ltd as a result of the substantive public and governmental discussions regarding concerns over acidic deposition and the need for immediate steps to address these concerns throughout Europe and in other countries. While there has always been a significant debate as to the primary causes and relative importance of different pollutants in the formation of acidic deposition and its damaging affects on sensitive eco-systems, there was general agreement in Europe that sulfur dioxide and nitrogen oxides originating from the combustion of fossil fuels in utility and industrial processes were a major contributor to the problem. A number of countries around the world embarked on major regulatory initiatives to reduce the emissions from stationary sources consuming fossil fuels. In Finland, the first steps toward identifying and developing appropriate environmental regulations addressing this problem were initiated in early 1985 through the work of a Sulfur Committee appointed by the Ministry for the Environment. It was in the subsequent development and promulgation of stringent regulations for controlling sulfur dioxide emissions at all existing, as well as new, coal-fired power plants in Finland that the LIFAC desulfurization process had its true beginnings.

---

<sup>1</sup> LIFAC is a registered trade mark of the process by Tampella Ltd. with a fully patented process approved in the U.S. in July of 1988.

As is commonly known, there were already many different processes in current commercial use on the market and Tampella itself had a long history in the successful manufacture and marketing of flue gas scrubbers (Figure 2). Particularly in the U.S., Japan, and the Federal Republic of Germany, development work and the commercial application of well known desulfurization processes had been in-place for several decades. The question arises then, why did Tampella depart from all of this experience and proven technology to pursue a new process, a new approach in the development of LIFAC?

While it was and is true that many wet and semi-dry desulfurization processes are very efficient and today are considered well proven technologies, the imposition of stringent environmental regulations on existing utility and industrial coal-fired facilities poses a whole new set of conditions, problems, and opportunities for new technologies. This was especially true for Tampella in that the approach to the problem was viewed very much from the perspective of the ultimate user -- desiring a simple, straightforward, reliable, yet economical system -- that could satisfy the new regulations being imposed as a result of concerns over acidic deposition. While the existing technologies were indeed efficient, the processes in fact are complex chemical processes requiring water treatment, pumping of washing liquids or sorbent slurries, difficult end product handling of the wet sludge from wet scrubbing systems, etc. Moreover, these processes required significant space and difficult design conditions that made the use of these technologies at existing power plants, in some cases, impossible and in others, excessively expensive. There appeared to be a clear need in the market for a user-friendly, simplified desulfurization process that was sufficiently efficient to meet the new stringent environmental regulations.

After a detailed study of the existing desulfurization methods, Tampella concluded that the possibility did exist for the development of a new process that would satisfy these requirements. To accomplish its research and product goals in light of the aggressive regulatory program established in Finland, and elsewhere, the following program objectives were identified for Tampella's development work:

- The complete development project had to be completed within 2 to 3 years including full-scale testing in order that the resulting process would be available at the time utilities would be making decisions regarding environmental compliance in Finland (compliance with the more stringent emissions limits had to be by 1993).
- The development work was to be accomplished primarily with the Company's own resources although cooperation with various universities and research institutes was also to be pursued.
- The end product of the development effort was to be a simplified and cost-effective desulfurization process, with total investment costs that would be roughly half those of the existing semi-dry and wet processes.
- The process should yield a waste product that is entirely water free and that this "dry" process should be readily applicable to both existing and new utility power plants and industrial processes.
- The process should yield high levels of SO<sub>2</sub> emission reductions irrespective of the combustion technique employed by the boilers, the fuel used or its sulfur content. Note that the minimum reduction requirements for the Finnish regulations were



approximately 80% SO<sub>2</sub> removal at existing power plants and 85% at new units.

#### THE IDEA BEHIND THE PROCESS

From the initial studies performed by Tampella of the existing processes, it was concluded that calcium-based processes were the most common. This appeared to be due to the ready availability and relative reasonable costs of limestone as well as the fact that limestone is easily handled and stored, either in raw form or as a pulverized product.

Limestone injection methods had already been studied in the 1950's and 1960's when mixing limestone with the coal feed to a boiler's pulverizers was tried. Since the limestone was pulverized with the coal and fired jointly into the boiler, the method was very simple requiring no separate equipment for feeding limestone into the furnace or for pulverization.

Low efficiency became, however, a stumbling block for the effective application of the method as a means of significantly reducing sulfur dioxide emissions. Only about 20-30% of the sulfur dioxide contained in the flue gases were removed and even to achieve these modest emission reduction results the amount of limestone required was quite large. Moreover, when boiler operators attempted to use large quantities of limestone problems arose involving slagging and fouling in the boiler furnace and superheater zones. This was the result of not only the large quantities of limestone added, but also the affect (often unpredictable) of the limestone on the melting behavior of the ash in the coal.

The development of limestone or sorbent injection became of interest again with the development and deployment of low-NO<sub>x</sub> combustion techniques. These techniques began to emerge towards the end of the 1970's. Through the application of these techniques it was possible to drop the temperatures in the boiler furnace, especially adjacent to the coal burner nozzles. Not only was this intended to improve or reduce NO<sub>x</sub> formation, it was believed that the lower temperatures would also reduce the risk of sintering the limestone particles injected into the furnace thereby reducing their reactivity in capturing SO<sub>2</sub> gases. Subsequent work in this area proved relatively unsuccessful in significantly improving sulfur dioxide removal efficiencies due to the varying temperature profiles within boiler furnaces and the variations in temperatures during boiler load changes.

It became clear that if limestone injection was to be successful, it would have to be done at elevations within the boiler that permitted maximum reaction with the sulfur dioxide in the flue gases while at the same time minimizing sintering. In order to determine the optimum temperature for limestone injection, both internal work was carried out at Tampella as well as reviewing the results of a large number of studies in Europe and in the U.S., which all generally indicated that temperatures between 850°C to 1000°C were optimum. In fluidized bed conditions, 850°C was found to be optimum, but in the fluidized bed technique the particle sizes and the retention times of limestone were found to be quite different than those require for pulverized coal applications. In the fluidized bed, particle sizes were approximately 500 µm and the retention times on the order of several minutes: Whereas, in the pulverized coal-fired boilers, the particle size tended to be less than 100 µm and the retention time in the desulfurization window of 800-1200°C on the order of 1 to 3 seconds.

The first laboratory tests by Tampella showed that in the pulverized coal-fired boiler the optimum feeding temperature for the limestone into the furnace

was more than 1000°C. Full-scale tests were carried out at a 150 Mwe pulverized coal-fired boiler at the Tampella Inkeroinen mill during 1984. While Tampella was able to determine optimum firing positions for injecting limestone into the boiler, even at its best, the SO<sub>2</sub> removals achieved were below 50% when limiting the limestone feed to reasonable quantities (Ca/S molar ratios of 3.0 or less). The result was that overall utilization of the calcium feed in desulfurization was only 15-20% and Tampella was far from its goal of achieving high levels of SO<sub>2</sub> removal.

Tampella began to struggle with the problem of how to increase the utilization of the limestone. One possibility investigated was the recirculation of the fly ash with relatively high quantities of unreacted limestone back into the boiler's furnace. It was soon determined that this approach was fairly impractical because of the unacceptably high limestone/ash loadings within the boiler resulting in slagging and fouling problems in the superheater zone of the boiler and difficulties with electrostatic precipitator performance.

Finally, Tampella began to explore the possibility of improving the utilization of the unreacted calcium oxide (CaO) from the boiler injection of limestone by "chemically" treating this material to encourage additional sulfur dioxide capture. Continuing with Tampella's objectives to keep the system simple and cost-effective, the idea of activating the unreacted calcium oxide leaving the boiler in a separate "activation reactor" using water was explored. The concept was that the calcium oxide would react with water to form calcium hydroxide (Ca(OH)<sub>2</sub>) which would readily react with the remaining SO<sub>2</sub> in the flue gases to form calcium sulfite (CaSO<sub>3</sub>).

Pursuing this concept in laboratory and bench scale testing proved very successful and the process began to emerge as a plausible concept which would meet the objectives established early on in the development phase by Tampella. The process was named LIFAC -- Limestone Injection into the Furnace and Activation of the unreacted Calcium. These successful preliminary tests led Tampella to advance to pilot scale and eventually to full-scale testing programs.

## TESTING

### Pilot Scale Testing

After additional literature studies and preliminary tests, the newly devised process of LIFAC was tested at the pilot scale level. Limestone injection had already been previously tested by Tampella in a 14MW<sub>th</sub> fluidized bed boiler and 150 MW<sub>e</sub> pulverized coal-fired boiler and so full-scale injection tests were continued with the LIFAC concept added on as a pilot scale test.

In cooperation with Pohjolan Voima Oy, full-scale limestone injection equipment and a pilot-scale activation reactor installed after the boiler's air preheater were retrofitted at the pulverized coal-fired boiler (220 MW<sub>e</sub>) in Kristiinankaupunki.

Due to Tampella's earlier experiences in testing the proper placement of limestone injection ports in boiler furnaces, the injection of limestone into the Kristiinankaupunki plant was raised to the level of its superheaters. At this placement, where the temperature is lower, overburning of limestone particles was minimized, thereby ensuring plenty of reactive material for sulfur removal within the activation reactor. Mild limestone firing conditions make binding sulfur dioxide in the furnace conditions possible, although not optimum, but on the other

hand it ensures optimum conditions for the post-reactions with water and sulfur dioxide in the after-reactor.

Attention in the pilot tests was primarily concentrated on testing limestone injection and developing preliminary information for modeling the operation of the activation reactor.

The results of the limestone injection confirmed that, as shown by the earlier experience, limestone sintering has to be prevented by feeding the absorbent at as low a temperature as possible within the boiler furnace, yet high enough to ensure calcination of the injected limestone. The LIFAC activation tests carried out by the side-stream reactor after injection were very promising (Figure 3), and so building a full-scale LIFAC plant was started at Imatran Voima's Inkoo Power plant. In addition, several other efforts were pursued to more fully understand and characterize the LIFAC process.

As the furnace injection of limestone is the major driving force of the entire LIFAC desulfurization process, it was decided in 1985 to participate in the project initiated by the fuel and lubricant division of the Technical Research Centre of Finland, the energy department of the Ministry of Trade and Industry, Imatran Voima and Partek. The project dealt with the applicability of different alkalis to desulfurization. The experiments were carried out at the drop tube furnace at the Technical Research Centre of Finland (Appendix 1).

At the same time, a LIFAC desulfurization system was built at the Nest Kulloo combustion laboratory as an add-on to an 8 MW<sub>th</sub> multifuel test boiler. The purpose of this test facility and development program was to extensively explore the relationships of the LIFAC process to key parameters.

The Kulloo trial runs made it possible to formulate the overall process behavior of LIFAC by an equation as follows:

$$X(\text{SO}_2) = f(C(\text{SO}_2), T_r, \Delta t, \text{Ca/S} \dots)$$

On the basis of the formulated equation depicting the process relationships, it is possible to present approximate estimations of the overall system behavior of LIFAC. Based on the values of the empirical test results, the function can be developed into an appropriate form which is easy to solve. The SO<sub>2</sub>-conversion ( $X(\text{SO}_2)$ ) is high, when the SO<sub>2</sub>-conversion ( $C(\text{SO}_2)$ ), gas retention  $T_r$  and Ca/S molar ratio are high and the temperature difference relative to the dew point ( $\Delta T$ ) of the flue gas is small. In Figure 4, the values calculated according to the above-mentioned reactor model are compared to the values observed in the many tests performed at the Kulloo test facility.

#### Full-Scale Testing at Producing Plants

The first full-scale LIFAC-desulfurization plant intended for continuous operation was built at Imatran Voima's Inkoo Power plant in 1985. This is a 250 MW<sub>e</sub> coal-fired power plant with seasonal cycling and seasonal full load operating characteristics. The limestone injection equipment was constructed for the full 250 MW<sub>e</sub> boiler and a LIFAC activation reactor was installed in one of two 125 MW<sub>e</sub> flue gas ducts coming from the boiler to two separate ESPs. A temporary ash silo was also built adjacent to the plant for collecting the end product for further examination.



The first stage testing of the process was concentrated on developing key insights into the reactions at the different stages of the LIFAC process; i.e., injection, reactor, and end product.

The objectives of the injection study were:

- to find out boiler fouling, if any, in different ways of operation;
- to establish the impact of injection on boiler availability and energy economy; and
- to clarify the  $\text{SO}_2$  level and conversion of calcium with different grades of limestone and Ca/S molar ratios.

The objectives of the reaction study were:

- to check the design parameters, equipment construction and material choices; and
- full-scale process testing with different  $\text{SO}_2$  levels.

The end product studies were intended to find out the following:

- the variations of the product quality as a function of different levels of operation;
- the ability to separate the process and boiler ash with the electrostatic precipitator without major modification;
- feasibility of storing the end waste products at landfill or dumping areas; and
- possibility of successfully utilizing in the end product in some commercial manner.

The tests showed that boiler fouling does not increase due to the limestone injection as designed by Tampella. Further, boiler availability does not appear to be impaired by the proper injection of limestone into the furnace either. Moreover, while ESP performance was degraded with only limestone injection, the addition of the activation reactor brought the overall ESP performance to well within the original emission levels without requiring modification.

The activation reactor helps to increase the  $\text{SO}_2$  removal by means of water injection, as specifically designed in the LIFAC process. When approaching the dew point temperature of the flue gas (or as the relative water amount approaches the value of 1, Figure 5) the sulfur reduction rate exceeds 80% at relatively moderate limestone loading levels ( $\text{Ca/S} < 2.0$ ).

Because of the successful results of the pilot tests at the first LIFAC reactor at Inkoo's 250 MW<sub>e</sub> power plant and at the Kulloo test boiler; in June 1987 Imatran Voima placed an order for an additional LIFAC reactor intended for continuous operation to control the sulfur dioxide in the flue gases at Inkoo's unit No. 4. The reactor was commissioned at the beginning of 1988, after which it has been used successfully in support of the overall normal operations of the plant. The process performance has been verified by several tests, and the emission level of 230 mg.  $\text{SO}_2/\text{MJ}$  set by the Finnish Government for all existing coal-fired plants of the size of Inkoo has been reached with Ca/S-molar ratios of



2.0 or less and a flue gas outlet temperature after the reactor of 65°C. This emission limit corresponds to about an 80% reduction in the SO<sub>2</sub> in the flue gases.

The emission level set by the Government for new plants was also demonstrated as attainable with the LIFAC process by raising the molar ratio through the injection of more pulverized limestone and/or by lowering the flue gas temperature after the reactor by means of water injection. The overall reduction in the SO<sub>2</sub> emissions of between 80-90% (see Figure 5) have been readily achieved. The sulfur content of the coal used at Inkoo has been 1.3 to 1.5%.

A number of studies have been made of the applicability of the end product for storage at landfill sites. The end product, due to its alkaline nature, decreases the dissolubility of the heavy metals in fly ash and the calcium sulfite contained in the product is stable. A large number of chemical analyses, column tests and agitation tests have been made at Imatran Voima's central laboratory on the end product material. These tests show that no harmful metals dissolve and the values remain well below the stringent limits set for drinking water in Finland (Appendix 2).

Storing at landfills, filling of excavations and roads, and applicability to road bases were studies conducted by Tampella in field tests of the end product. The behavior of the product was examined by different methods in a number of different applications, particularly with respect to possible groundwater contamination. All of these tests confirmed the ability to landfill or use the end product in construction type applications with no difficulties.

#### **ECONOMY**

The costs of the flue gas desulfurization are highly dependent on the plant size, site characteristics and conditions, limestone costs, the level of SO<sub>2</sub> reductions required, and the process chosen. The process costs are especially important when considering wet or spray-drying processes.

The investment costs of the LIFAC process consist of limestone injection equipment, reactor automation and control systems, and ash handling equipment. In operating coal-fired boilers, the particle collector most often can be the original electrostatic precipitator or the baghouse already installed at these existing boilers.

As in the all FGD-processes, the consumption of the absorption chemical or sorbent constitutes the highest operating cost in the LIFAC process.

In Table 1 the costs of the LIFAC process are calculated for existing coal-fired generating facilities for selected plant sizes. The calculations shown reflect 1988 values and estimate levelized total costs in terms of constant 1988 Finnish marks.

#### **SUMMARY**

Tampella's LIFAC-desulfurization process is currently a commercial process which has been under development since 1984. The duration of the research and development work, the conversion of the initial concepts into a practical method, has been very short; i.e., only four years. Active and efficient cooperation with universities and research institutes has greatly assisted in this development effort and the ability to fully commercialize a new process in so short a period

of time. Typically, developments of this nature require a decade or more to move the process from the initial conceptual stage to a commercial product.

The laboratory and pilot-scale tests have made it possible for Tampella to create design criteria and modeling of the critical elements and reactions for the process. Tampella has the ability to reliably dimension the process into full scale applications for a wide range of conditions and fuel use. Tests at the Kulloo test boiler using Pittsburgh 8 seam coal have confirmed the performance of the LIFAC process for American high-sulfur bituminous coals.

The main results achieved at the full scale plants are:

- verification of criteria
- verification of technical reliability
- confirmation of reliable, continuous operation
- confirmation of the basis of mathematical modeling and process design parameterization
- verification of the suitability of the end product for landfill disposal
- suitability of the end product for use as a landfill for embankments and probably for road base material.

New LIFAC deliveries now under contract will occur during this next year, 1989, when the LIFAC system will be delivered to Vantaa electricity board (Finland) and to a pulp and paper factory in Baikal (Soviet Union). Tampella is actively seeking a utility partner for demonstration of its LIFAC process in the U.S. as part of a Clean Coal III project.

#### DISCLAIMER

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

FIGURE 1  
The LIFAC Process Developed by Tampella Ltd's Boiler Division

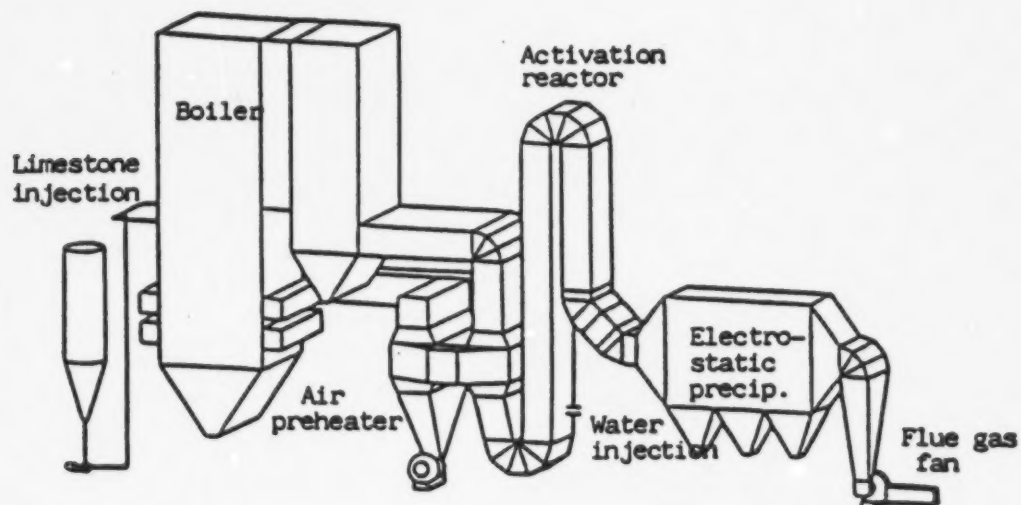


FIGURE 2  
Tampella Flue Gas Scrubber

**Tampella**  
BOILER DIVISION

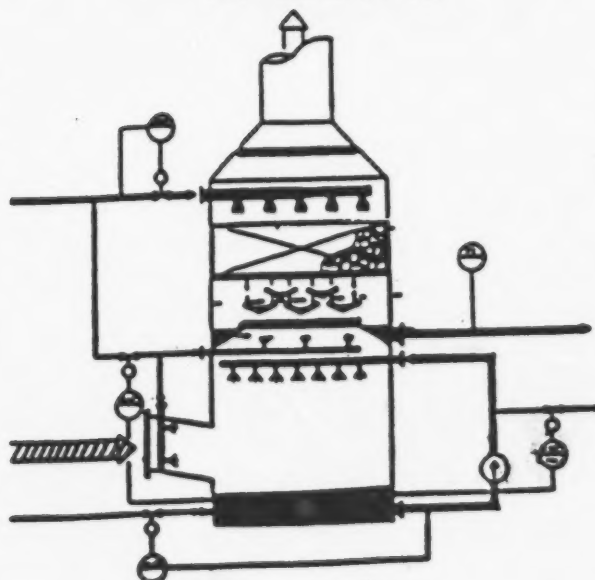


FIGURE 3  
Change in the  $\text{SO}_2$  Level  
Preliminary Tests, Kristiinankaupunki, 1985

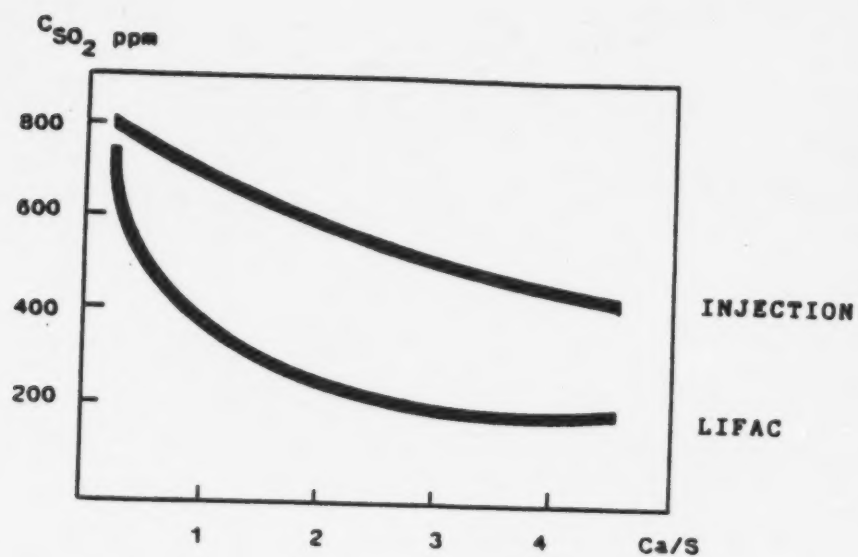


FIGURE 4  
Correlation of the Reaction Model with the Empirical Results

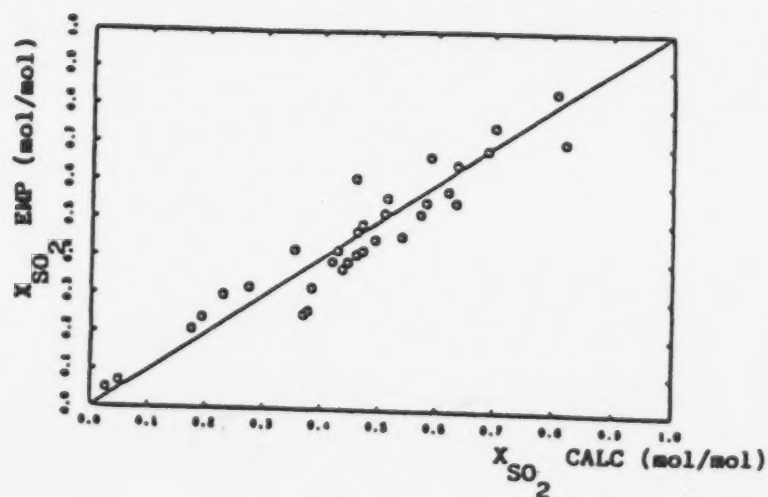




FIGURE 5  
 $\text{SO}_2$  Reduction Rate at the LIFAC Process

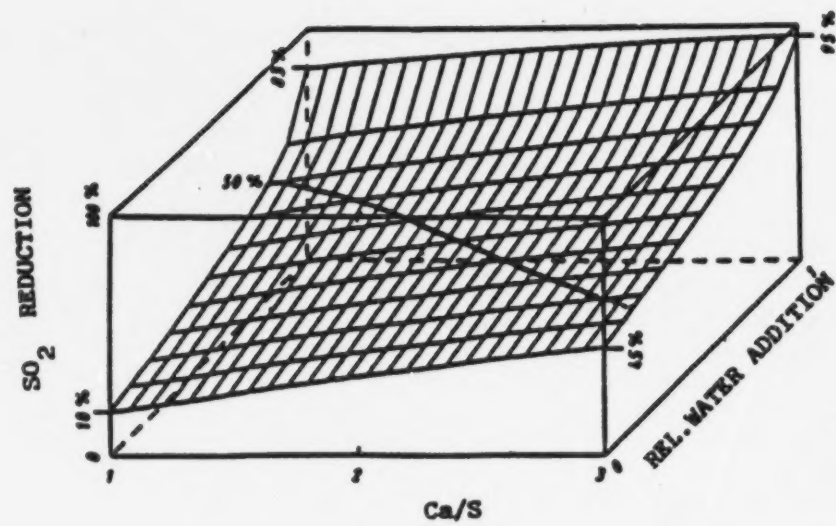


TABLE 1

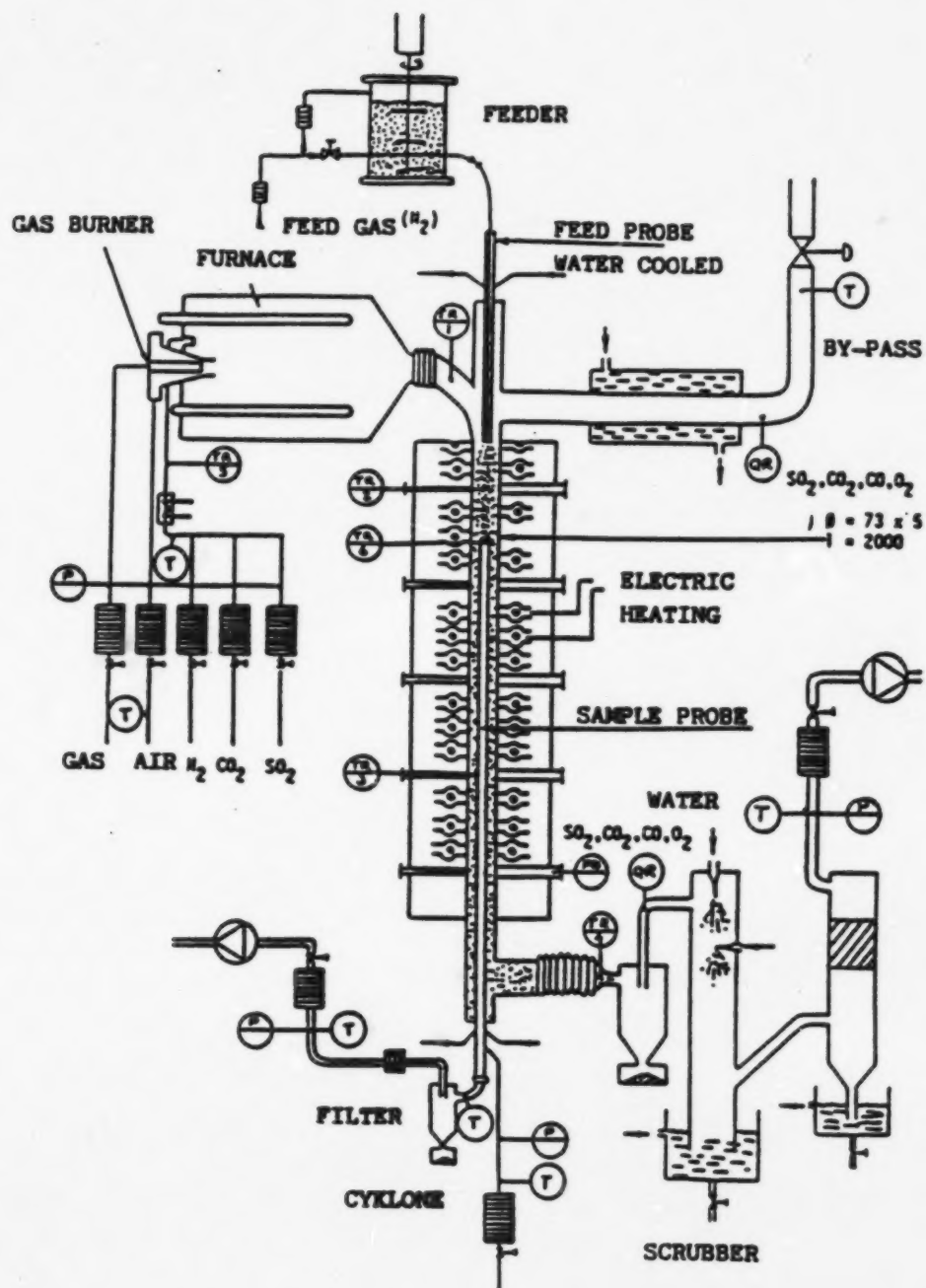
## The Overall Operating and Capital Costs of the LIFAC Process

| Powerplant Size                                           | 340 MW <sub>e</sub> |              | 500 MW <sub>e</sub> |
|-----------------------------------------------------------|---------------------|--------------|---------------------|
| SX                                                        | 1.0                 | 3.2          | 3.2                 |
| SO <sub>2</sub> Reduction Rate                            | 80%                 |              | 80%                 |
| Investment FIM/kW <sub>e</sub>                            | 200...240           | 200...250    | 190...240           |
| Operating Cost<br>(FIM/MWh <sub>e</sub> )                 |                     |              |                     |
| - Limestone                                               | 3.7                 | 11.8         | 11.8                |
| - Other oper.                                             | 3.7                 | 4.9          | 4.9                 |
| Investment Costs<br>(FIM/MWh <sub>e</sub> ) <sup>1/</sup> | 2.4                 | 2.4          | 2.3                 |
| Total Costs<br>FIM/MWh <sub>e</sub>                       | 9.4                 | 19.1         | 19.0                |
| Approximate<br>US\$/MWh <sub>e</sub> <sup>2/</sup>        | -----<br>2.4        | -----<br>4.8 | -----<br>4.8        |

1/ Assumes 6,000 hours of operation (approximately 68 percent capacity factor and annualizing the investment costs at 25 years with a 5% real discount.

2/ Roughly divide total Finnish Marks by four to estimate U.S. dollar equivalent.

# APPENDIX 1 Drop Tube Furnace



APPENDIX 2  
Column and Agitation Tests with the End Product

|                        | <u>Drinking Water in Finland</u> <sup>1</sup> | <u>Column Test</u> <sup>2</sup> | <u>Agitation Test</u> <sup>3</sup> |
|------------------------|-----------------------------------------------|---------------------------------|------------------------------------|
| pH                     | 6.0 - 9.0                                     | 12.0                            | 12.0                               |
| NO <sub>2</sub> (mg/l) | 0.2 - 1.0                                     | <0.1                            | <0.1                               |
| NO <sub>3</sub> (mg/l) | 30 - 50                                       | 5                               | 0.8                                |
| SO <sub>4</sub> (mg/l) |                                               | 350                             | 200                                |
| PO <sub>4</sub>        |                                               | <0.1                            | <0.1                               |
| Fe                     | 0.3 - 1.0                                     | 0.86                            | 0.24                               |
| Mn                     | 0.1 - 0.5                                     | 0.04                            | <0.04                              |
| Cu                     | 0.3 - 1.0                                     | 0.06                            | 0.01                               |
| Pb                     | 0.05                                          | <0.05                           | <0.05                              |
| Cd                     | 0.005                                         | <0.01                           | <0.01                              |
| Zn                     | 1.0 - 3.0                                     | 0.12                            | 0.01                               |

1 The Quality of Drinking Water IN Finland, National Board for Health, 1985

2 Ratio solution-solids 0.1

3 Ratio solution-solids 1:1



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### SORBENT SLURRY INJECTION FOR SO<sub>2</sub> CONTROL

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by: M.S. Mozes, R. Mangal and R. Thampi  
Ontario Hydro Research Division  
800 Kipling Avenue  
Toronto, Ontario  
M8Z 5S4  
Canada

#### ABSTRACT

In-furnace sorbent slurry injection for controlling SO<sub>2</sub> emission from power plant flue gases has been studied under conditions simulating full scale operation at Ontario Hydro's 640 MJ/h Combustion Research Facility. These studies are an important component of the corporation's acid gas control program and are aimed at optimizing SO<sub>2</sub> capture, sorbent utilization and ESP performance with sorbent injection. Two calcitic limestone slurries from Ontario and one from Nova Scotia were tested. Coals tested include an eastern US bituminous coal, a blend of an eastern US and a western Canadian coal and a Nova Scotia coal. Injection temperature, residence time, slurry concentration and stoichiometry and mode of injection have been optimized to maximize SO<sub>2</sub> capture and sorbent utilization and to minimize the impact on particulate removal by the ESP. The results indicate that sulphur capture is a sensitive function of injection parameters (quality of atomization, discharge momentum of the droplets and distribution and mixing with the flue gas). Under optimized operating conditions while burning a 1.7% S US bituminous coal and injecting a 40% Pt. Anne limestone slurry cocurrently with the flue gas at 1200°C, up to 70% SO<sub>2</sub> can be removed at a Ca/S ratio of 2.5 to 3.0. SO<sub>2</sub> capture and sorbent utilization by the other limestone slurries was slightly lower. Ash resistivities are up by about two orders of magnitude yet in a range where ESP performance is good.

#### INTRODUCTION

Ontario Hydro is committed to reduction of the level of acid gas emission from its coal burning plants. The main pollutants of concern are sulphur dioxide and nitrogen oxides. In December, 1985, the Ontario Government announced stricter acid gas emission limits for Ontario industries and Ontario Hydro's limits were reduced to 430,000 tonnes/year starting in 1986 and 215,000 tonnes/year starting in 1994. Nine nuclear units coming into service will help to reduce coal burns and hence gas emissions.

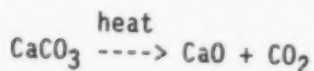
Burning low sulphur coals is another option being considered to reduce Ontario Hydro's acid gas emissions into the early 1990's. In the 1990's uncertainties in load forecasts and the demand/supply options indicate that wide variation in coal burns are possible and various control technologies may be required (1).

To ensure that economic options are available to enable Ontario Hydro to meet the emission limits, a major program is underway to improve acid gas control by sorbent injection. This process could have a role in Ontario Hydro's acid gas control scenarios for a number of reasons. The technology is less capital intensive than FGD, therefore it is likely to have a cost advantage for boilers with short remaining life; it is ideally suited for retrofit application and it requires shorter lead times for installation than wet scrubbing systems.

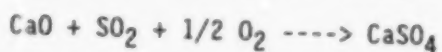
The schematic diagram of the slurry injection process is shown in Figure 1a. In this process an aqueous spray of Ca-based sorbent is injected into the combustion chamber at temperatures ranging between 990 to 1200°C. The technique provides improved distribution and mixing of the sorbent with the gas flow, prevents sorbent deactivation and allows for sufficient residence time at favourable temperatures for the reaction between SO<sub>2</sub> and CaO to be efficiently completed (2).

The following steps shown in Figure 1b are involved in the process:

- o Sorbent-water atomization and intimate mixing with flue gas; water droplets evaporation; particle disintegration due to increased vapour pressure in the pore structure;
- o Calcination resulting in additional fragmentation, development of high surface area and pore structure. As the water evaporates the sorbent is calcined and forms very fine and well dispersed particles, ready to react with SO<sub>2</sub> in a temperature range of 990 to 1200°C. At these temperatures sintering, loss of calcine surface area and reactivity is very slow.



- o Sulphur capture by the porous reactive sorbent in a favourable thermal environment.



- o The CaSO<sub>4</sub> and unreacted CaO are collected with the fly ash in the electrostatic precipitator.

The most important variables which influence process performance are: sorbent dispersion, determined by the nozzle performance; injection location, coal and sorbent characteristics; and sorbent slurry particle size, concentration and injection rate. Optimization of the above variables under conditions simulating full scale operation was carried out for maximum sulphur capture and sorbent utilization using a 1.7% sulphur US

bituminous coal and an Ontario limestone. A 1.5% eastern US/W Canadian coal blend was also tested. The process was also evaluated for another Ontario limestone with the 1.7% S Coal and a 3% sulphur Nova Scotia Devco coal with a Nova Scotia limestone.

## EXPERIMENTAL

### COMBUSTION RESEARCH FACILITY

The study was conducted at Ontario Hydro's Combustion Research Facility (CRF) designed for a maximum coal feed rate of about 20 kg/h (44 lb/h) US bituminous coal at a firing rate of 640 MJ/h ( $6.1 \times 10^5$  BTU/h) (Figure 2). The furnace is a refractory-lined cylindrical chamber, fully equipped for monitoring gas and wall temperatures. There are multiple ports for flame observation and for insertion of solid sampling probes. There are also probes to determine slagging and fouling rates. The pulverized coal is delivered down-draft to the burner with the combustion air which can be electrically preheated to temperatures up to 350°C (662°F). Gas burners on each side of the coal burner are used to heat the furnace to operating temperatures before beginning to feed to coal.

The coal burner, designed and constructed by Research Division staff, is equipped with a vortex generator and four air vanes to assure good mixing and adequate residence time of the fuel-air mixture in the combustion zone. The combustion gases in the furnace are cooled by water and/or air circulating in the cylindrical inconel jacket around the furnace. This cooling system is equipped with temperature sensors and flow meters to control furnace quenching rates.

The combustion gases leaving the furnace are further cooled by a series of air-cooled heat exchangers prior to entering the resistivity probe housing and ESP. The ESP consists of a cubic stainless steel chamber, and is equipped with two sets of interchangeable cells. One set has an 11-plate electrode with 2.5 cm (1 in) spacing, the other a 5-plate electrode with 5 cm (2 in) spacing. The design specific collection areas (SCA,  $m^2/m^3/s$ ) for the two sets of cells are 39 (0.2 ft<sup>2</sup>/cfm) and 17 (0.09 ft<sup>2</sup>/cfm) respectively for baseline firing conditions using a high volatile US bituminous coal.

The CRF instrumentation permits system temperatures, and flue gas compositions (O<sub>2</sub>, CO<sub>2</sub>, CO, SO<sub>2</sub> and NO<sub>x</sub>) to be monitored continuously. Gas temperatures in the furnace are measured with a suction pyrometer and flame temperatures with an optical pyrometer. Flow rates and pressures are measured by flow meters and manometers. All measuring and monitoring systems are linked to a computerized data acquisition system. Particulate mass loading in the flue gas before and after the ESP is measured with an isokinetic sampling system. In-situ resistivity is measured with a point-plane resistivity probe situated in the resistivity probe housing and particle size distribution of the ash is measured with a Pilot Mark 3 Cascade Impactor.

A more complete description of the facility is given elsewhere (3).

## SLURRY INJECTION SYSTEM

A slurry injection system, shown in Figure 3 was designed and successfully commissioned. A positive displacement pump pumps the slurry from a continuously stirred mixing tank under a pressure of 650 to 720 kPa. Slurry recirculation and a static mixer upstream of the furnace keep the particles in suspension and prevent settling. A small metering pump delivers the slurry to the atomizer through which fine droplets are injected into the flue gas stream.

The slurry is injected into the middle of the furnace through a twin-fluid high pressure nozzle (5 or 3 mm) with an internal mixing chamber, shown in Figure 4. Operating pressures range between 40 to 60 psig. The stainless steel nozzle was purchased from Turbotak Inc. The MMD of the droplets are about 14  $\mu\text{m}$  for the 5 mm nozzle and approximately 9  $\mu\text{m}$  for the 3 mm nozzle. The nozzle is equipped with a cooling jacket which was necessary to avoid evaporation of the water and hence drying of the slurry causing deposition of particles in the nozzle. In order to maximize sulphur capture and sorbent utilization it was recognized that proper mixing of the slurry with the combustion gases was essential. Furnace temperature probing with a suction pyrometer was carried out at different elevations of the furnace to make sure that severe stratification in combustion gas temperature profiles did not exist. Injecting the slurry at the middle of the furnace was found to be best and  $\text{SO}_2$  capture was found to be a function of droplet size which in turn is directly related to the atomizing air pressure.

## PROCEDURES

After steady state was achieved with the baseline coal, injection of the sorbent slurry into the middle of the furnace at specified locations ( $I_1$  to  $I_4$ ) was initiated. Temperature-time and radial profiles of flue gases under conditions simulating Lakeview and Lambton TGS thermal profiles are shown in Figure 5. Data collected during each test include system temperatures, and pressures, slurry-feed rates and stoichiometry, flue gas constituents concentrations ( $\text{CO}_2$ ,  $\text{O}_2$ ,  $\text{CO}$ ,  $\text{SO}_2$  and  $\text{NO}_x$ ) and in-situ ash resistivities. Coal, sorbent feed and fly ash samples were collected during the tests. Analysis of samples include chemical composition and particle size distribution. Calcines and sulphated calcines were analyzed for  $\text{CaO}$ ,  $\text{CaCO}_3$  and  $\text{CaSO}_4$  content. Porosity, mass median diameter and BET surface area of some samples were also determined. The analytical methods used are described in reference (3).

A high volatile, medium sulphur eastern US coal from Lakeview TGS, a medium sulphur blend of W. Canadian and Eastern US coals and a high sulphur coal from Nova Scotia were used in the study. The proximate and ultimate analyses and particle size distribution of the coals are given in Table 1.

Sorbents used in the study include: two local calcitic limestones and a Mosher Irish Cove limestone from Nova Scotia. Chemical and physical properties of these sorbents are given in Table 2.



## RESULTS AND DISCUSSIONS

The parameters that were found to affect sulphur capture efficiency include:

Sorbent composition, porosity, particle size and surface area;

Injection parameters;

Temperature at the point of injection;

Slurry concentration, and

Ca/S ratio.

### SORBENTS COMPARISON

Sorbent composition and physical properties are the most important factors in determining overall capture performance (4,5,6,7,8,9). Significant variability in the reactivity of the various sorbents has been observed as shown in Tables 3, 4 and Figure 7. Pt. Anne limestone with an initial porosity of 55% gave significantly higher removal than Beachville limestone with an initial porosity of 17% (70% removal for Pt. Anne compared to 55% for Beachville). The Nova Scotia limestone slurry was used with the Nova Scotia coal. Thus a direct comparison of process performance between this sorbent and the local calcitic stones was not possible. Data indicate however, that similar sulphur capture can be obtained with Nova Scotia limestone and the Pt. Anne limestone even if they are used for two different coals. Porosity of both limestones 55%-57%; sulphur content of the corresponding coals is 1.7% and 2.8%.

### EFFECT OF INJECTION PARAMETERS

Injection parameters that influence  $\text{SO}_2$  capture include: atomizer type, injector location, atomizing air pressure, and particle size distribution or mass median diameter (MMD) of the atomized droplets. High atomization air pressure improves the quality of atomization and promotes an early release of the sorbent particles to engage in the sulphation reaction. It also increases the discharge momentum of the droplets leading to enhanced penetration and mixing with the gas stream.

The effect of varying atomizing air pressure is shown in Figure 6.  $\text{SO}_2$  capture was found to be a function of the atomizing air pressure which in turn is directly related to droplet size distribution, quality of atomization and mixing with the flue gas. The best mixing was observed while spraying a 40% aqueous Pt. Anne limestone slurry into the furnace cocurrently at injection location  $I_2$ , which was close to the flame zone where increased turbulence exists. Increasing the atomizing pressure from 40 psig to 70 psig improves the  $\text{SO}_2$  capture from about 62% to 70% at Ca/S ratio of 3.0.

## EFFECT OF TEMPERATURE AND INJECTION MODE

The effect of temperature on  $\text{SO}_2$  capture was evaluated for the Pt. Anne limestone slurry and Beachville limestone slurry while burning a 1.7% S eastern US bituminous coal. A 1.5% eastern US/western Canadian coal blend was also evaluated with the Pt. Anne slurry. Both the cocurrent and countercurrent injection modes at Ca/S ratio of 3.0 were investigated. The results are given in Table 3 and plotted in Figure 7.

The performance of the 3 mm and 5 mm nozzles was also compared. The 3 mm nozzle, which produces finer droplets than the 5 mm one performs much better at both injection modes for the Pt. Anne limestone slurry tested with the US coal. Cocurrent injection results in slightly higher sulphur capture than countercurrent injection at the higher temperature zone. If the slurry is injected at lower temperatures ( $975^\circ\text{C}$ ), countercurrent injection is preferred. At about  $1100^\circ\text{C}$  both injection modes result in similar  $\text{SO}_2$  capture.

The best  $\text{SO}_2$  removal, about 70%, was obtained by injecting the Pt. Anne slurry through a 3 mm nozzle at  $1200^\circ\text{C}$ , cocurrently into the furnace while burning the US coal. With countercurrent injection the optimum temperature is  $1100^\circ\text{C}$  with an  $\text{SO}_2$  capture of about 66%. Injecting the Pt. Anne slurry cocurrently with the coal blend at  $1200^\circ\text{C}$  resulted in a maximum  $\text{SO}_2$  capture of 66% while countercurrent injection at  $1100^\circ\text{C}$  gave the best capture of 63%. It is interesting to note that for the cocurrent injection mode, sulphur removal within the range of  $1350$  to  $1100^\circ\text{C}$  is not a very sensitive function of the temperature. This is a very useful feature of the process, implying that temperature fluctuations at  $1200^\circ\text{C}$  in the full scale might not detrimentally affect process performance.

The cocurrent injection of a 40% Beachville limestone slurry with an initial porosity of 17% shows only a 55%  $\text{SO}_2$  capture with the US coal. At the countercurrent injection mode lower temperatures are preferred. At  $1150^\circ\text{C}$  both modes of injection results in similar  $\text{SO}_2$  removal.

## EFFECT OF SORBENT STOICHIOMETRY

The effect of Ca/S ratio on sulphur capture with the US coal was evaluated at  $1200$  and  $1300^\circ\text{C}$  for the Pt. Anne limestone slurry, and at  $1200^\circ\text{C}$  for the Beachville limestone.

Evaluation of the coal blend was done at  $1200^\circ\text{C}$  with the Pt. Anne limestone slurry. The results are shown in Table 4 and plotted in Figure 8.

As the data indicate, sulphur capture decreases with decreasing Ca/S ratios, but the drop is not as sharp as with dry sorbent injection. Even at a ratio of 1.5 for the Pt. Anne slurry, 50%  $\text{SO}_2$  can be captured at  $1200^\circ\text{C}$  and 48% at  $1300^\circ\text{C}$  while burning the US coal. The less porous Beachville limestone shows 31% removal at a ratio of 1.5. Calcium utilizations are 33 and 21% respectively. This is probably due to the fact that at lower stoichiometries, slurry mass flow rate to the atomizer is reduced while maintaining high atomizing air pressure. This results in

improved atomization, penetration and mixing of the slurry with the flue gas and hence improved process performance.

#### EFFECT OF RESIDENCE TIME

The effect of residence time on  $\text{SO}_2$  capture in the sulphation zone, (1200 - 900°C) was studied by varying furnace quenching rate. Changing the quenching rate from 500°C/s (simulating Lakeview TGS quenching rate) to 280°C/s (simulating Lambton TGS quenching rate) increases the residence time from 600 ms to 900 ms at an injection location of 1200°C. No change in  $\text{SO}_2$  capture was observed from the 70% capture observed at 600 ms while burning a 1.7% S US coal and injecting a 40% Pt. Anne slurry at a Ca/S ratio of 3.0 cocurrently into the furnace. One explanation is that with the injection of an aqueous spray of a calcium based sorbent, porous, reactive calcine is formed. This in turn gives rise to a high rate of calcium utilization in the furnace in a very short time. The sulphation reaction is then rapidly terminated, thus increased residence time does not improve future sorbent utilization.

#### EFFECT OF COAL

Three coals were studied during the test program. A medium sulphur (1.7%) eastern US bituminous coal, a high sulphur (2.8%) bituminous coal from Nova Scotia tested with a Nova Scotia Mosher limestone and a 1.5% S coal blend of an eastern US bituminous and western Canadian coals. Unfortunately because of impurities (grit) in the Nova Scotia limestone and limited pump capacity, only a 20% slurry at a ratio of 2.2 could be fed. This complicates a direct comparison of process performance with the US coal and coal blend where a 40% aqueous slurry of Pt. Anne limestone was sprayed at a Ca/S ratio of 3.0. However, it did offer an opportunity to show that maximum  $\text{SO}_2$  capture peaked at the same injection temperature of 1200°C for all coals and that similar trends in  $\text{SO}_2$  removal versus injection temperatures were observed as shown in Figure 9.

It is interesting to note that injecting the Pt. Anne limestone slurry at a ratio of 2.5, with the US coal resulted in 60%  $\text{SO}_2$  capture, comparable to  $\text{SO}_2$  capture by the Nova Scotia limestone slurry at a ratio of 2.2 with a 2.8 % sulphur Nova Scotia coal. Injecting the Pt. Anne slurry with the coal blend gave 55%  $\text{SO}_2$  removal at a ratio of 2.5. These data indicate that high  $\text{SO}_2$  capture can be achieved by slurry injection with high sulphur coals.

#### IMPACT ON ASH CHARACTERISTICS, COLLECTABILITY AND DEPOSITION

During sorbent slurry injection fly ash is intimately mixed with reaction products (mainly  $\text{CaSO}_4$ ) and unreacted sorbent (mainly  $\text{CaO}$ ). With such waste some unanswered questions regarding sorbent slurry injection are its impact on fly ash characteristics, precipitability, and deposition in the radiant section (slagging) and convective passes (fouling) of a boiler. Chemical composition, particle size distribution and carbon-content of the waste are also important aspects for design considerations for waste management systems. The effect on the

back end of sorbent slurry injection was studied and the results are given below.

#### Ash Characteristics

In Table 5, particle size distribution of isokinetically collected ash samples from the injection of various sorbent slurries are compared to the baseline ash. Particle size distribution is plotted in Figure 10.

The mass median diameter of the baseline ash is about  $8\text{ }\mu\text{m}$  compared to  $6\text{ }\mu\text{m}$  for the Pt. Anne slurry waste and  $9\text{ }\mu\text{m}$  for the Beachville slurry waste. The waste from the Pt. Anne Slurry is finer than that of the baseline. This is expected since the mass median diameter of the Pt. Anne limestone is  $3.9\text{ }\mu\text{m}$ . The data indicate that particle size distribution will not affect particle migration velocity and hence particle size distribution does not appear to be a problem for the ESP during slurry injection.

In Table 6 a typical waste from slurry injection is compared to the baseline ash and to a waste from dry sorbent injection. High levels of calcium compounds and the quantity produced still remains a problem for handling and disposal. CaO content of a typical slurry waste is 269 g/kg and  $\text{CaSO}_4$  content is about 232 g/kg. The waste produced is about twice that of the baseline ash and the ESP's SCA may need to be increased to handle the extra burden during sorbent slurry injection.

#### Dust Electrical Resistivity And ESP Performance

The resistivity of the baseline fly ash measured in-situ with about 23 ppm  $\text{SO}_3$  naturally occurring in the flue gas from the medium sulphur eastern U.S. bituminous coal is about  $10^8\text{ ohm}\cdot\text{cm}$ . During injection of all slurries, resistivities consistently increased by one to two orders of magnitude to  $10^9$  to  $10^{10}\text{ ohm}\cdot\text{cm}$  yet the electrical operating conditions of the ESP were not severely affected and collection efficiencies were not seriously degraded (see Table 7) from a baseline level of 89% during slurry injection. Dry injection on the other hand results in a resistivity of  $10^{11}\text{ ohm}\cdot\text{cm}$  and an 8% drop in collection efficiency.

It is possible that due to the increased moisture level in the flue gas (up to 23% relative humidity) a thin film of water forms around the particles and acts as a conditioning agent aiding the ESP in its performance.

Inlet mass loading to the ESP has increased 2 to 3 fold from a baseline level of  $1.4\text{ g/m}^3$  with a resulting increase in particulate emissions by a factor of about 3 times during slurry injection. Thus the main problem with slurry injection is the high dust loading to the ESP which depends on the Ca/S ratio.

#### Slagging And Fouling Propensities Of The Waste

Soft deposits, which form at low temperatures and are generally characteristic of deposits found on air heaters and economizers were observed on the furnace walls and heat exchanger surfaces. These deposits could be easily blown away by compressed



air suggesting that conventional soot blowing equipment may suffice for full scale application of slurry injection.

#### CONCLUSIONS

In-furnace injection of sorbent slurries is a more efficient way of removing  $\text{SO}_2$  from flue gases than dry injection. This technique facilitates improved distribution and mixing of the sorbent with the gas flow, reduces sorbent deactivation and allows sufficient residence time at favourable temperatures for the reaction between  $\text{CaO}$  and  $\text{SO}_2$  to be efficiently completed. In this study sorbent slurry injection of two calcitic limestones were used to remove  $\text{SO}_2$  from flue gases while burning a 1.7% S eastern US bituminous coal and a 1.5% S coal blend of eastern US and western Canadian coal, and a Nova Scotia Mosher limestone slurry was used to remove  $\text{SO}_2$  from a high sulphur (2.8%) coal from Nova Scotia. The results of the study conducted at Ontario Hydro's 640 MJ/h Combustion Research Facility are summarized below:

- o Sorbent properties are very important in determining the degree of sulphur capture available via slurry injection. Pt. Anne limestone with an initial porosity of 55% was superior to Beachville limestone with an initial porosity of only 17%.
- o Injection parameters were found to be also very important in capturing  $\text{SO}_2$ . High atomizing air pressure which improves the quality of atomization, promotes an early release of the sorbent and increases the discharge momentum of the droplets, increased  $\text{SO}_2$  capture significantly. The Turbotak 3 mm twin fluid atomizer produced the finest droplets and performed the best. Increasing the atomizing medium pressure from 40 to 70 psig increased  $\text{SO}_2$  capture from 62 to 70% at a Ca/S ratio of 3.0.
- o  $\text{SO}_2$  removal was found to be a sensitive function of injection temperatures and mode of injection. The optimum injection temperature for all limestone and coals tested was  $1200^\circ\text{C}$ . Injecting a 40% Pt. Anne limestone slurry cocurrently into the flue gas at  $1200^\circ\text{C}$  while burning a 1.7% S eastern US bituminous coal produced an  $\text{SO}_2$  capture of 70% at a Ca/S ratio of 3.0. Injecting a Beachville limestone slurry under similar operating conditions resulted in only a 55% removal.  $\text{SO}_2$  capture for the high sulphur (2.8%) Nova Scotia coal while injecting a 20% Nova Scotia limestone slurry at a Ca/S ratio of 2.2 was about 60%.
- o Residence time was found to have negligible effect on sulphur capture.

Increasing the residence time in the sulphation window from 600 to 900 ms by decreasing furnace quenching rate did not result in additional capture.

- o Sulphur capture for all coals and sorbents tested decreases but sorbent utilization increases with decreasing Ca/S ratios. At a Ca/S stoichiometry of 1.5 for the Pt. Anne slurry, while burning the US coal, sulphur capture dropped from 70% to 50%, but sorbent utilization increased from 23% to 33%. Similar trend was observed for the various coals-limestones.
- o Particle size distribution of the waste from the Pt. Anne slurry was slightly finer than the baseline ash. However, high levels of calcium compounds ( $\text{CaO}$ ,  $\text{CaSO}_4$ , etc.) and the quantity of waste produced remains a problem for handling and disposal.
- o Ash resistivities increased by one to two orders of magnitude from  $10^8$  ohm.cm to  $10^9$  to  $10^{10}$  ohm.cm but ESP collection efficiencies were not seriously affected. The increased level of the flue gas moisture is believed to act as a conditioning agent.
- o Slagging does not appear to be a problem and the soft deposit formed on the furnace walls and heat exchanger surfaces was easily removable.

#### FUTURE WORK

Studies are in progress on sorbent slurry injection where reduced spray droplet size and selected additives are being investigated to improve sulphur capture, sorbent utilization and ESP performance. Tests are also in progress to assess a series of additives to simultaneously control  $\text{NO}_x$  emissions. The reburning process for  $\text{NO}_x$  control will also be studied and integrated with sorbent injection. Waste activation and recycling and in-duct sorbent injection - humidification is also planned.

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the agency and no official endorsement should be inferred.

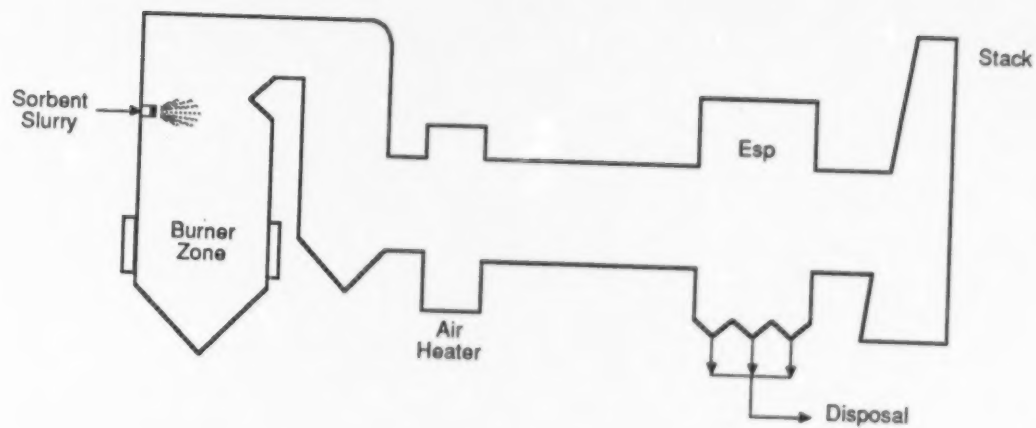
#### ACKNOWLEDGMENTS

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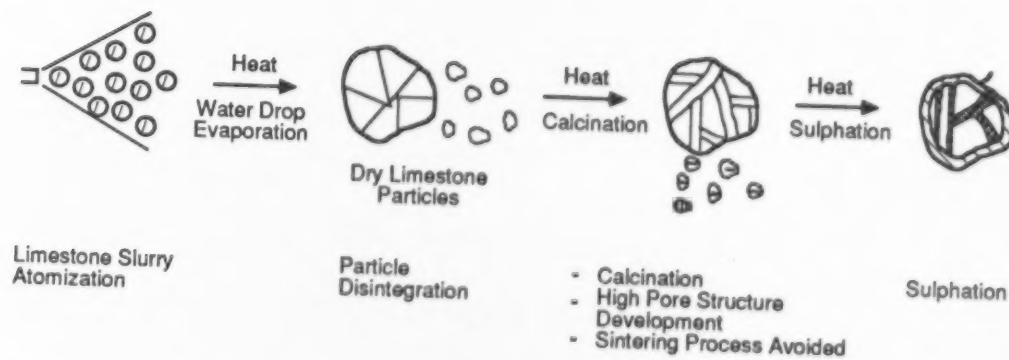
Design and Development Division. The authors wish to express a special thanks to Mr. H.A. Bamborough of Environment Canada, Mr. D.K. Blamire of Nova Scotia Power and Mr. C.W. Dawson of Ontario Hydro for their useful comments and suggestions during the test program.

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(a) Schematic Process Diagram



(b) Chemical and Physical Steps

Figure 1. Sorbent Slurry Injection



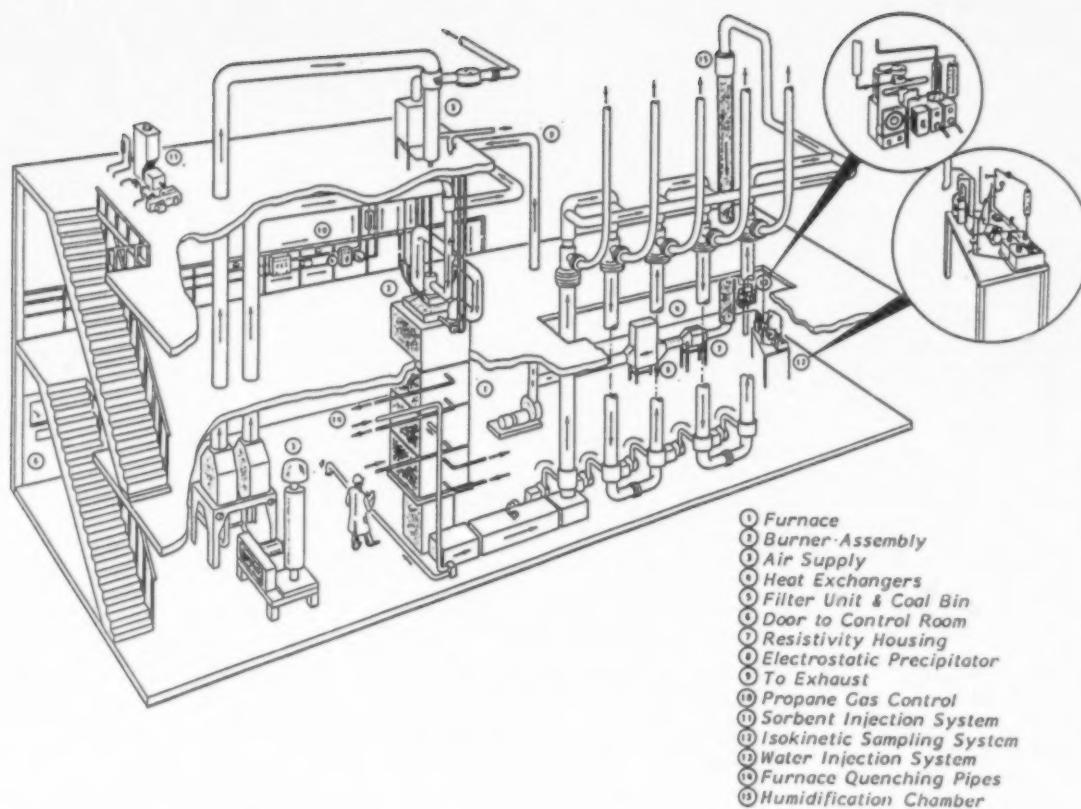


Figure 2. Combustion Research Facility

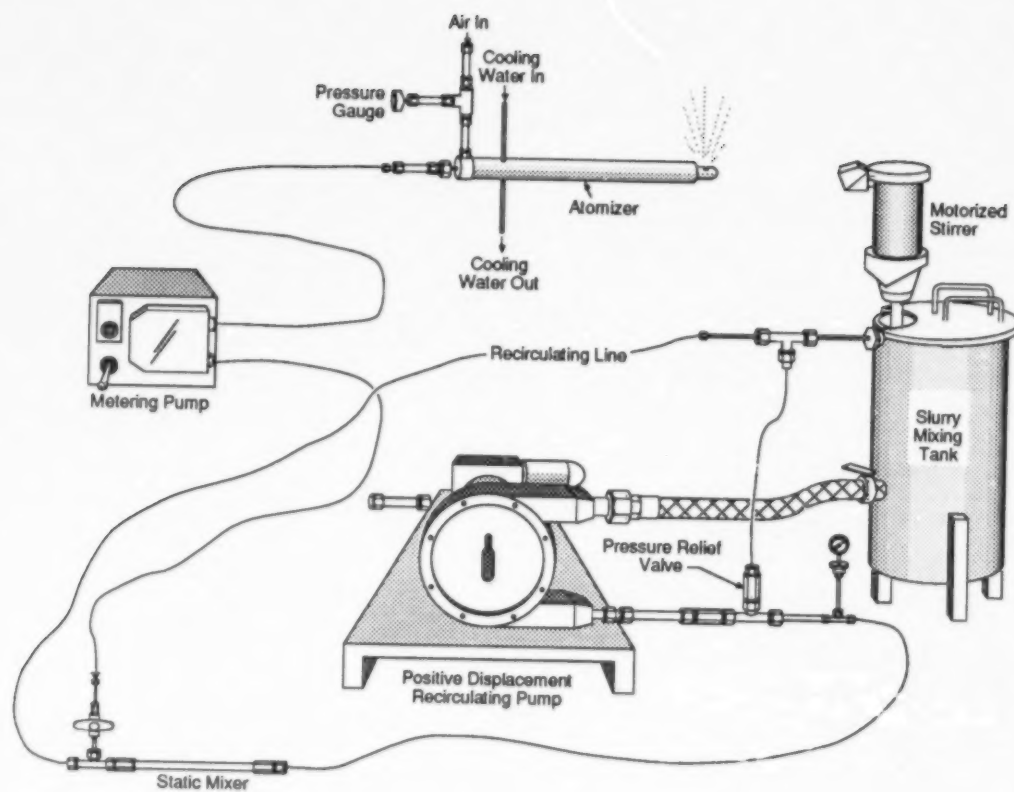


Figure 3. Slurry Injection System

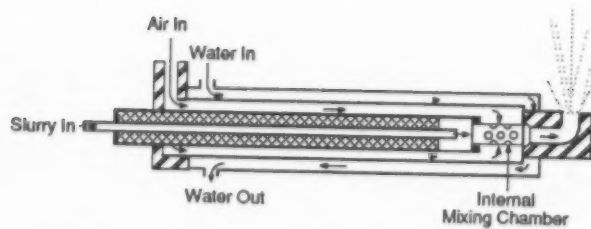


Figure 4. Turbotak "Extended" Nozzle

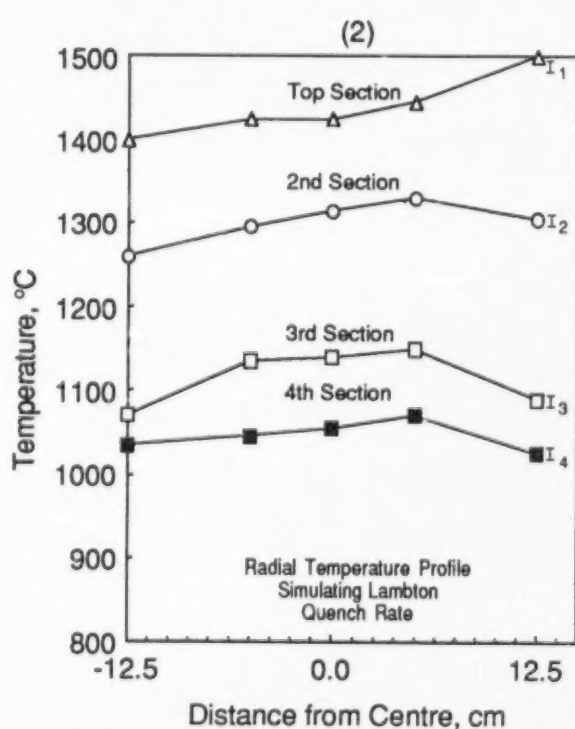
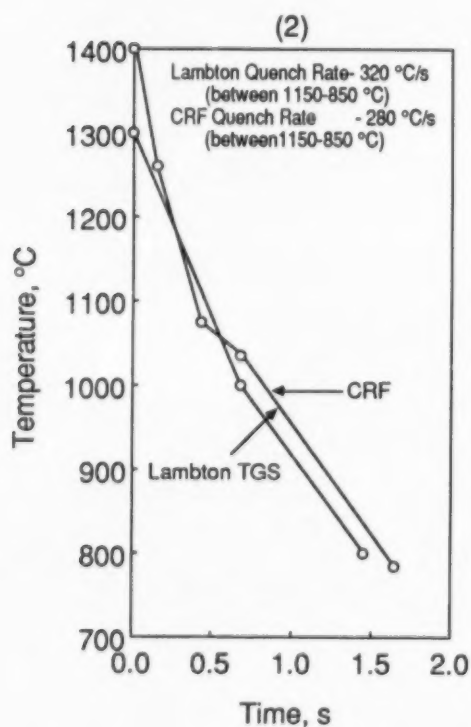
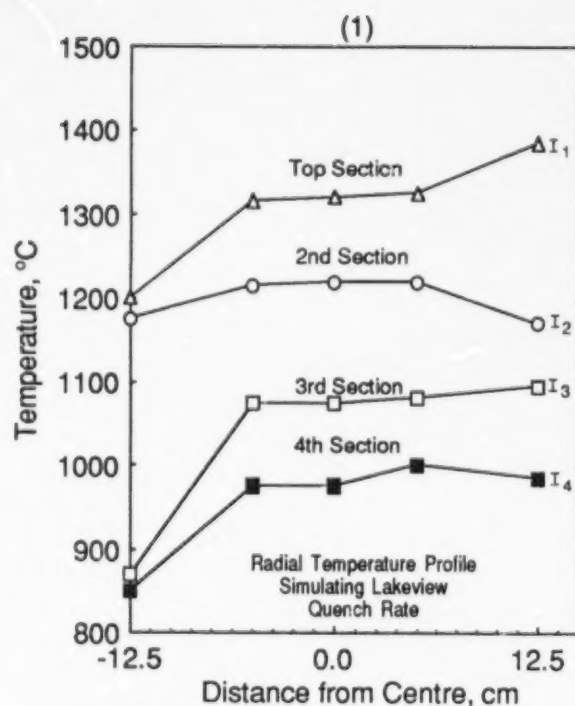
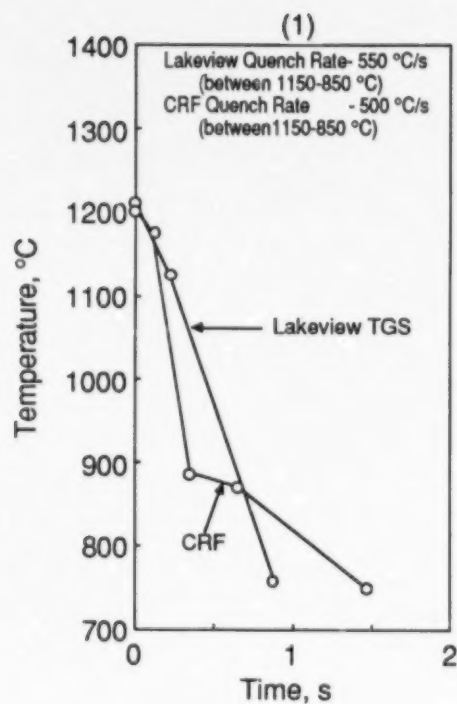


Figure 5. Temperature-Time and Radial Profiles of Flue Gases

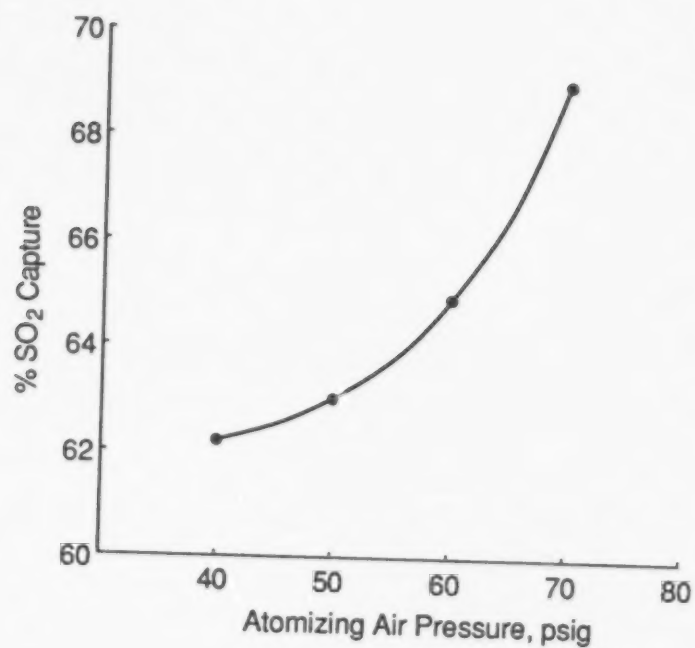


Figure 6. SO<sub>2</sub> Capture as a Function of Atomizing Air Pressure

Nozzle - 3 mm Turbotak  
40 % Aqueous Slurry of Pt. Anne  
Limestone, Cocurrent Injection  
Temp: 1200 °C  
Ca / S = 3.0



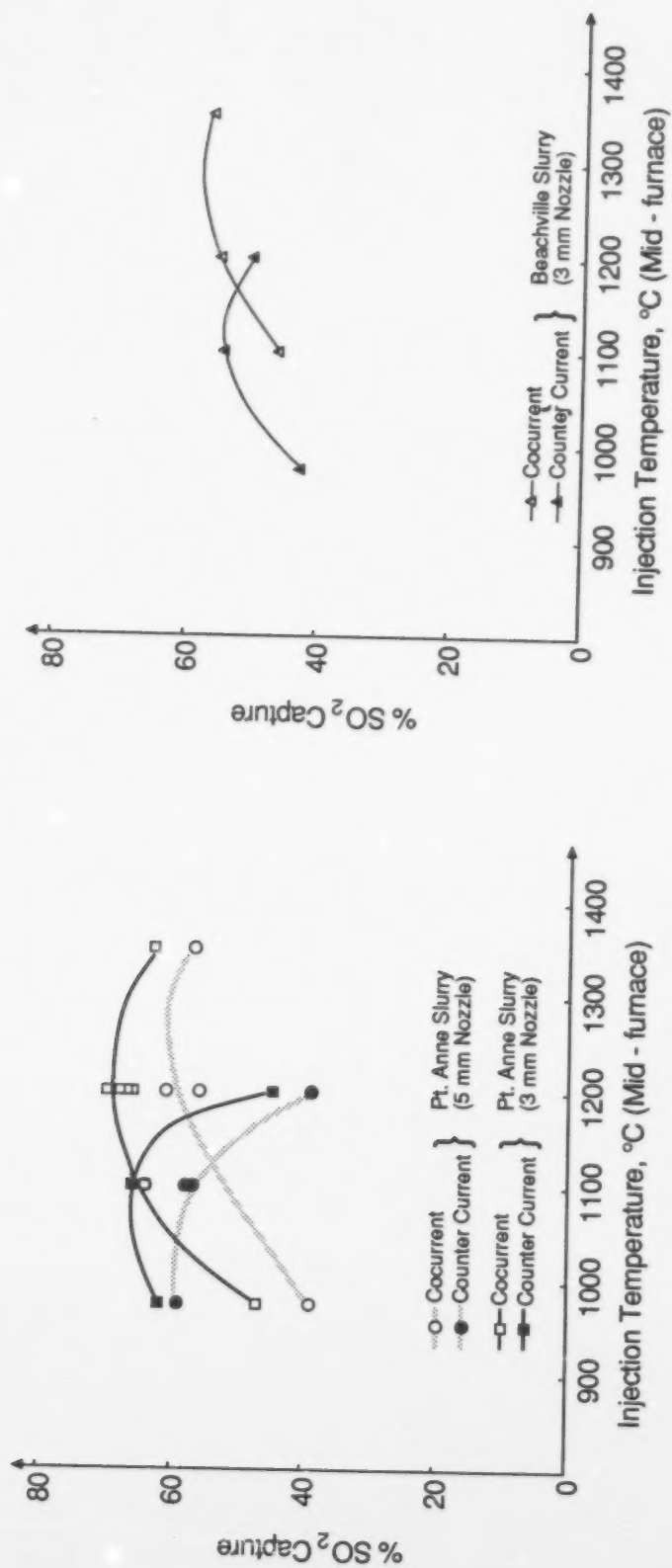


Figure 7. SO<sub>2</sub> Removal vs Injection Temperature

1.7% Eastern US Coal  
40% Aqueous Slurry of Pt. Anne Limestone  
Ca / S = 3.0

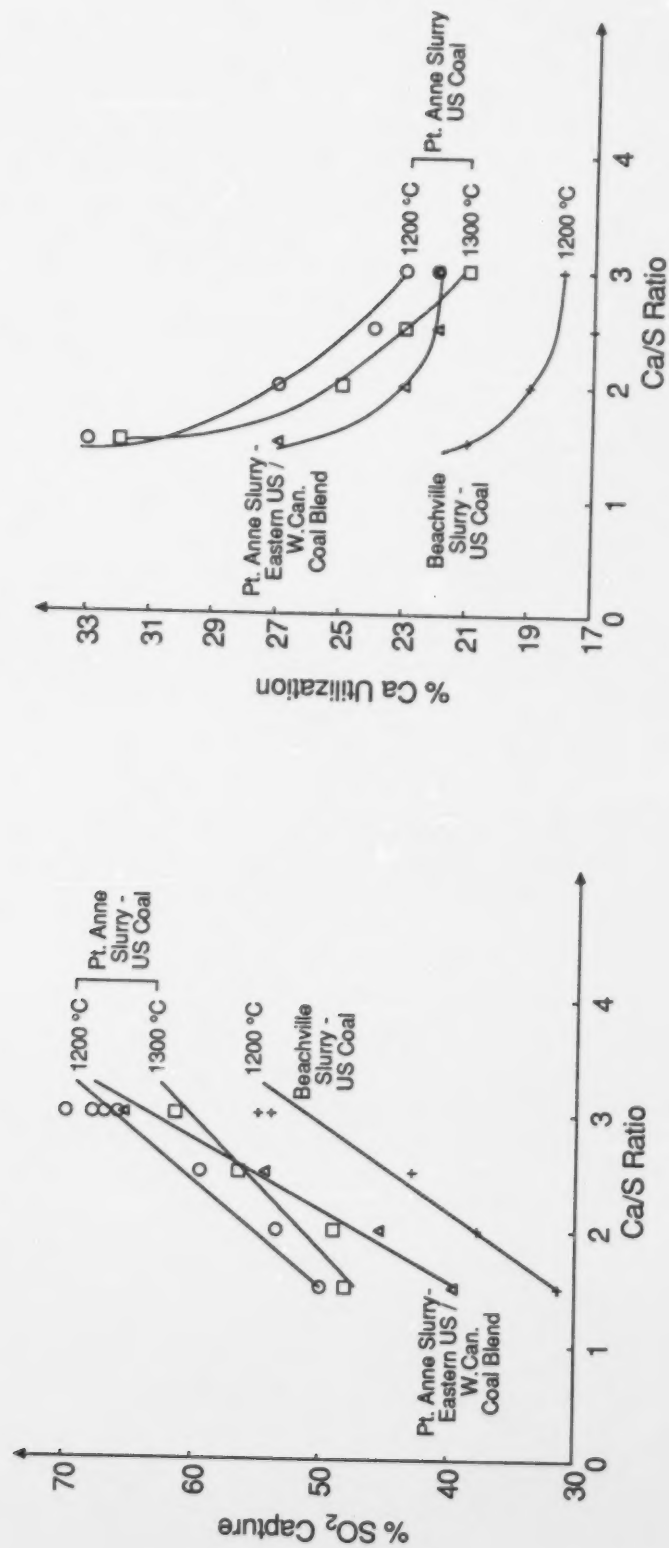


Figure 8. Effect of Ca / S Ratio

Nozzle - 3 mm Turbotak  
Atomizing Air Pressure 70 psig  
40 % Aqueous Slurry

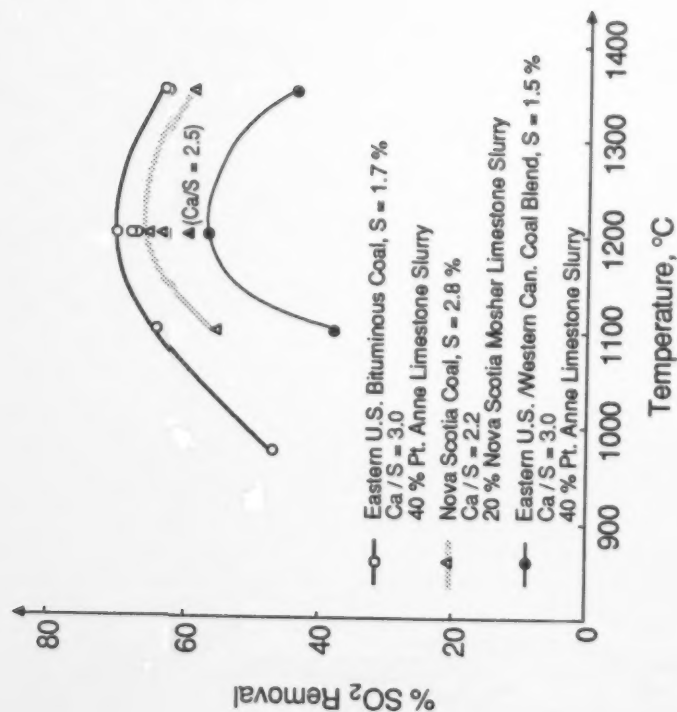


Figure 9. Effect of Coal

- 3 mm Nozzle  
- Co-Current Injection

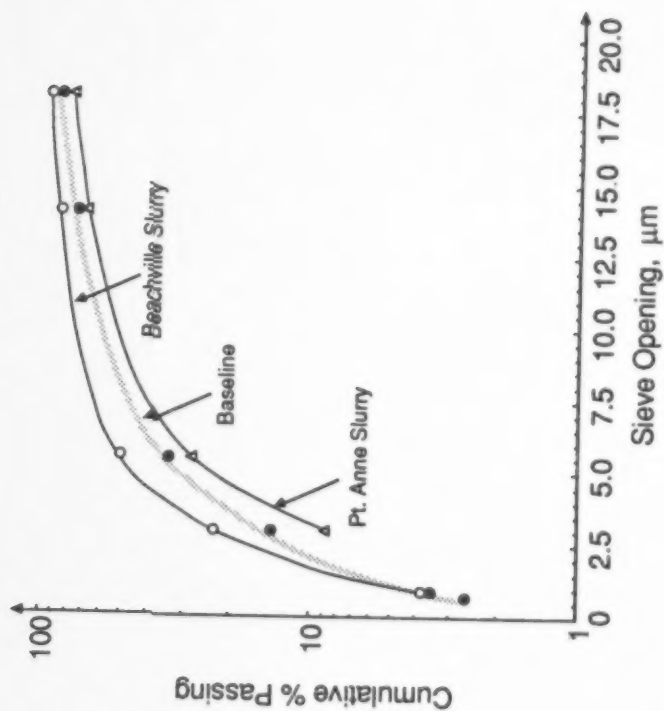


Figure 10. Particle Size Distribution of Baseline Flyash and Slurry Wastes

40% Slurry  
1.7% S Eastern US Coal  
Ca/S = 3:1

**TABLE 1**  
**CHARACTERISTICS OF COALS**

| Proximate Analysis, g/kg |         |                  |                     | Ultimate Analysis, g/kg |         |                  |                     |
|--------------------------|---------|------------------|---------------------|-------------------------|---------|------------------|---------------------|
|                          | US Coal | Nova Scotia Coal | W.Can-US Coal Blend |                         | US Coal | Nova Scotia Coal | W.Can-US Coal Blend |
| Moisture                 | 14      | 12               | 32                  | Carbon                  | 756     | 756              | 751                 |
| Ash                      | 80      | 96               | 99                  | Hydrogen                | 57      | 50               | 47                  |
| Volatile Matter          | 357     | 314              | 321                 | Nitrogen                | 16      | 12               | 11                  |
| Fixed Carbon             | 548     | 577              | 548                 | Sulphur                 | 17      | 28               | 15                  |
| Heating Value MJ/kg      | 32      | 31               | 29                  | Ash                     | 80      | 96               | 71                  |
|                          |         |                  |                     | Oxygen                  | 74      | 58               | 101                 |

**Particle Size Distribution**

| Sieve Opening $\mu\text{m}$ | Cumulative % Passing |                  |                     |
|-----------------------------|----------------------|------------------|---------------------|
|                             | US Coal              | Nova Scotia Coal | W.Can-US Coal Blend |
| 150                         | 100                  | 100              | 100                 |
| 106                         | 93                   | 100              | 98                  |
| 75                          | 73                   | 92               | 84                  |
| 63                          | 59                   | 84               | 76                  |
| 53                          | 56                   | 75               | 63                  |
| 44                          | 51                   | 49               | 19                  |
| <44                         |                      |                  |                     |

**TABLE 2**  
**CHEMICAL AND PHYSICAL PROPERTIES OF SORBENTS**

|                                 | Beachville Limestone | Pt. Anne Limestone | Mosher Limestone (Nova Scotia) |
|---------------------------------|----------------------|--------------------|--------------------------------|
| $\text{Li}_2\text{O}$ , g/kg    | <0.03                | -                  |                                |
| $\text{Na}_2\text{O}$           | 0.03                 | 0.1                | 0.1                            |
| $\text{K}_2\text{O}$            | 0.6                  | 0.5                | 1.1                            |
| $\text{MgO}$                    | 8.0                  | 4.8                |                                |
| $\text{CaO}$                    | 524.0                | 535.4              | 538.0                          |
| $\text{Fe}_2\text{O}_3$         | 0.1                  | 2.1                | 6.4                            |
| $\text{Al}_2\text{O}_3$         | 22.0                 | 4.3                | 6.5                            |
| $\text{SiO}_2$                  | <12.0                | 21.0               | 25.0                           |
| LOI                             | 434.0                |                    |                                |
| BET area, $\text{m}^2/\text{g}$ | 1.3                  | 2.9                | 1.86                           |
| MMD, $\mu\text{m}$              | 8.6                  | 3.9                | 11.0                           |
| $\rho$ , $\text{g}/\text{cm}^3$ | 2.6                  | 2.3                | 2.5                            |
| POROSITY, %                     | 17.0                 | 55.0               | 57.0                           |



**TABLE 4**  
**EFFECT OF Ca/S RATIO AND SORBENT UTILIZATION**  
40% Aqueous Slurry  
Co Current Injection  
Turbotak 3mm Nozzle

| Coal                   | Slurry     | Injection Temp. °C | Ca/S Ratio | % SO <sub>2</sub> Removal | % Sorbent Utilization |
|------------------------|------------|--------------------|------------|---------------------------|-----------------------|
| 1.7 % S US             | Pt. Anne   | 1200               | 1.5        | 50                        | 33                    |
|                        |            |                    | 2.0        | 53                        | 27                    |
|                        |            |                    | 2.5        | 60                        | 24                    |
|                        |            |                    | 3.0        | 65-70                     | 20-23                 |
| 1.7 % S US             | Pt. Anne   | 1300               | 1.5        | 48                        | 32                    |
|                        |            |                    | 2.0        | 49                        | 25                    |
|                        |            |                    | 2.5        | 56                        | 23                    |
|                        |            |                    | 3.0        | 67-70                     | 20-23                 |
| 1.5 % S US-W.Can Blend | Beachville | 1200               | 1.5        | 31                        | 21                    |
|                        |            |                    | 2.0        | 38                        | 19                    |
|                        |            |                    | 2.5        | 43                        | 17                    |
|                        |            |                    | 3.0        | 55                        | 18                    |
| 1.5 % S US-W.Can Blend | Pt. Anne   | 1200               | 1.5        | 40                        | 27                    |
|                        |            |                    | 2.0        | 45                        | 23                    |
|                        |            |                    | 2.5        | 54                        | 22                    |
|                        |            |                    | 3.0        | 66                        | 22                    |

**TABLE 5**  
**PARTICLE SIZE DISTRIBUTION OF BASELINE AND SLURRY INJECTION WASTES**

| Size $\mu$ m | Particle Size Distribution |                       |                         |
|--------------|----------------------------|-----------------------|-------------------------|
|              | Baseline Ash               | Pt. Anne Slurry Waste | Beachville Slurry Waste |
| 1            | 4                          | 4                     | 9                       |
| 3            | 14                         | 23                    | 24                      |
| 5            | 30                         | 47                    | 69                      |
| 14           | 74                         | 85                    | 79                      |
| 18           | 87                         | 91                    |                         |

**TABLE 3**  
**EFFECT OF TEMPERATURE AND INJECTION MODE**  
40 % Aqueous Slurry  
Ca/S Ratio = 3.0  
Lakeview TGS Quenching Rate

| Coal                                  | Slurry                          | INJECTION |                           | Nozzle Turbotak<br>Caldyn | % SO <sub>2</sub><br>Removal |
|---------------------------------------|---------------------------------|-----------|---------------------------|---------------------------|------------------------------|
|                                       |                                 | Location  | Temp. °C<br>(Mid Furnace) |                           |                              |
| 1.7 % S Eastern US Bituminous         | Pt. Anne                        | 1 1       | 1350                      | 5 mm                      | 57                           |
|                                       |                                 | 1 2       | 1200                      |                           | 36, 31                       |
|                                       |                                 | 1 3       | 1100                      |                           | 48                           |
|                                       |                                 | 1 4       | 975                       |                           | 39                           |
|                                       |                                 | 1 1       | 1350                      | 3 mm                      | 63                           |
|                                       |                                 | 1 2       | 1200                      |                           | 66, 65, 67, 68, 70           |
|                                       |                                 | 1 3       | 1100                      |                           | 64                           |
|                                       |                                 | 1 4       | 975                       |                           | 47                           |
|                                       |                                 | 1 2       | 1200                      | 5 mm                      | 39                           |
|                                       |                                 | 1 3       | 1100                      |                           | 64, 57, 58, 52, 51           |
|                                       |                                 | 1 4       | 975                       |                           | 59                           |
|                                       |                                 | 1 2       | 1200                      | 3 mm                      | 45                           |
|                                       |                                 | 1 3       | 1100                      |                           | 66                           |
|                                       |                                 | 1 4       | 975                       |                           | 62                           |
| 1.5 % S Eastern US-W.Can Blend        | Beachville                      | 1 1       | 1350                      | 3 mm                      | 57                           |
|                                       |                                 | 1 2       | 1200                      |                           | 55                           |
|                                       |                                 | 1 3       | 1100                      |                           | 46                           |
|                                       |                                 | 1 2       | 1200                      | 3 mm                      | 50                           |
|                                       |                                 | 1 3       | 1100                      |                           | 55                           |
|                                       |                                 | 1 1       | 1350                      | 3 mm                      | 59                           |
|                                       |                                 | 1 2       | 1200                      |                           | 63, 66                       |
|                                       |                                 | 1 3       | 1100                      |                           | 56                           |
|                                       |                                 | 1 4       | 975                       |                           | 53                           |
|                                       |                                 | 1 2       | 1200                      | 3 mm                      | 63                           |
|                                       |                                 | 1 3       | 1100                      |                           | 57                           |
|                                       |                                 | 1 4       | 975                       |                           | 41                           |
| 2.8 % S Nova Scotia Moshier Limestone | * Nova Scotia Moshier Limestone | 1 1       | 1350                      | 3 mm                      | 44                           |
|                                       |                                 | 1 2       | 1200                      |                           | 58, 63, 59                   |
|                                       |                                 | 1 3       | 1100                      |                           | 37                           |
|                                       |                                 | 1 4       | 975                       |                           | 41                           |

\* Ca / S = 2.2

**TABLE 6**  
**TYPICAL WASTE COMPOSITION**

Ca/S= 2.5  
SO<sub>2</sub> Removal = 62%  
40% Limestone Slurry  
Injection Temperature = 1200°C (mid-furnace)

|                          | Baseline<br>g/kg | * Sorbent Slurry<br>Injection Waste<br>g/kg | Dry Injection<br>Waste<br>g/kg |
|--------------------------|------------------|---------------------------------------------|--------------------------------|
| CaO                      | 34               | 269                                         | 281                            |
| CaSO <sub>4</sub>        | 39               | 232                                         | 178                            |
| CaCO <sub>3</sub>        | 30               | 40                                          | 90                             |
| MgO                      | 9                |                                             |                                |
| LOI                      | 47               | 143                                         | 143                            |
| kg Waste/<br>100 kg Coal | 8.8              | 17                                          | 16                             |

\*Waste normalized to 143 g/kg LOI

**TABLE 7**  
**SORBENT SLURRY INJECTION**  
**BACK END STUDIES:**

Inj Temp 1200°C (mid furnace)  
40% Limestone Slurry

| Slurry       | Ca/S<br>Ratio | % SO <sub>2</sub><br>Removal | % Utilization | % Relative<br>Humidity | Ash<br>Resistivity<br>ohm-cm | % LOI | ESP Performance                               |                                              |                 |
|--------------|---------------|------------------------------|---------------|------------------------|------------------------------|-------|-----------------------------------------------|----------------------------------------------|-----------------|
|              |               |                              |               |                        |                              |       | Dust<br>Loading<br>Before<br>g/m <sup>3</sup> | Dust<br>Loading<br>After<br>g/m <sup>3</sup> | Efficiency<br>% |
| Baseline Ash |               |                              |               | 8 - 10                 | $8.3 \times 10^7$            | 4.7   | 1.4                                           | 0.15                                         | 89              |
| Beachville   | 3.0           | 55                           | 18            | 23                     | $4.7 \times 10^9$            | 3.9   | 3.0                                           | 0.36                                         | 88              |
| Pt. Anne     | 2.5           | 62                           | 25            | ~19                    | $1.1 \times 10^{10}$         | 5.0   | 3.7                                           | 0.42                                         | 87              |
| Beachville   | 3.0 (Dry)     | 43                           | 14            | 8 - 10                 | $1.1 \times 10^{11}$         | 9.5   | 5.6                                           | 1.1                                          | 81              |

Desulfurization Results  
for Three Different Firing Concepts

by: Dr. M. Yaqub Chughtai  
L. & C. Steinmüller GmbH  
Gummersbach FRG

FIRST COMBINED FGD AND DRY SO<sub>2</sub>  
CONTROL SYMPOSIUM

ENVIRONMENTAL  
PROTECTION  
AGENCY

&

ELECTRIC POWER  
RESEARCH  
INSTITUTE

St. Louis, Missouri  
October 25.-28....1988

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## 1. ABSTRACT

After a short general description of the direct Desulfurization process, the commercial application to three different firing concepts namely stoker, ignifluid and pulverized fuel is described. Desulfurization results along with operating experience on the boiler ancillary equipment from different plants are discussed in detail. Different solutions for the disposal of by-products practiced by plant owners are presented and discussed.

The further development of direct Desulfurization will concentrate on the better utilization of the sorbent. Two different processes are described, one concerned with fly-ash conditioning and recirculation and the other with flue gas conditioning. As the results show, the combination of these processes makes direct Desulfurization more attractive for commercial applications.

## 2. GENERAL

Protection of our environment is one of the most important basic requirements for improving the quality of life in modern industrial societies. In the Federal Republic of Germany this objective is implemented by, among other regulations, the Bundesimmissionsschutzgesetz (federal anti-pollution law) and the TA-Luft (technical instructions on air pollution control). These laws and directives are designed to limit the emissions of airborne pollutants. It is expected that these limits, especially those of the TA-Luft, will be successively tightened up over the next few years.

Special attention has been paid to sulphur dioxide emissions and it is regarded as extremely important that these be reduced as far as possible.

Oxides of sulphur are released in huge quantities during the combustion of fossil fuels and in a great variety of industrial processes. It is possible to reduce these sulphur dioxide emissions by cleaning the flue gases before they are given off to the atmosphere.

For smaller firing systems in particular, simple and inexpensive processes are in great demand.

To ensure that the environmental problem is solved and not merely transferred from the air to the water resources, the residues from the flue gas cleaning plants must ideally be transformed into usable by-products which have an assured market for many years to come.

## 3. DIRECT DESULFURIZATION PROCESS

Steinmüller very early on began to develop plants and plant components to enable firing systems to comply with legal emission requirements and also addressed its efforts to the particular problem of developing a Desulfurization process especially for small firing systems.

This background and Steinmüllers intimate knowledge of the requirements firing systems operators place on their equipment have made it possible to develop an efficient, economical and flexible system for use in almost any type of equipment.

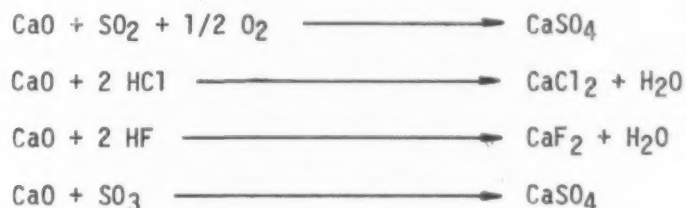
The basis of our system is the direct Desulfurization process (DDP). Under DDP we understand the chemical capture of sulphur within the boiler furnace itself.

The method of direct Desulfurization discussed here is absorption of sulphur by the basic sorbents  $\text{CaCO}_3$  (powdered limestone) and  $\text{Ca(OH)}_2$  (slaked lime). Fig. 1 shows the qualitative process of sulphur capture, plus that of any halogens present.

When  $\text{CaCO}_3$  or  $\text{Ca(OH)}_2$  are employed as the additive for pollutant capture, endothermic reaction decarbonation or dehydration (calcination) takes place in the furnace in accordance with the following reactions:



The CaO particles react with sulfur oxides and other pollutants like halogens according to the following exothermic reactions.



As a result of the direct Desulfurization process the additive particles are covered with a compact sulphate layer. Material transport of the  $\text{SO}_2$  deep into the pores and adequate diffusion velocity in the boundary layer can only be attained at the cost of very long residence times (minutes or hours), i.e. the core of the additive particle is never available for combination with  $\text{SO}_2$ .

In order to make this free CaO nevertheless available for further sulfur capture, there are two different methods which have been developed by Steinmüller. The one is of flue gas conditioning and the other of fly-ash conditioning and recirculating.

The precondition of applying these additional processes for better sorbent utilization is the presence of free active CaO in the flue gas coming out of the boiler.

For accomplishing the flue gas conditioning process, the water is atomized in the quench cooler which evaporates, and encouraged by the hygroscopic properties of the free CaO, it diffuses within the pores of a CaO core surrounded by a layer of CaSO<sub>4</sub>. The free CaO in the pores then combines with the H<sub>2</sub>O to form Ca(OH)<sub>2</sub>. This exothermic reaction tends to progress outwards from the pores on the exterior of the particle and is also encouraged by discontinuities and substitution molecules. This is termed the primary surface increase.

Since the Ca(OH)<sub>2</sub> has a lower bulk density and a greater specific surface area than the CaO, the resultant expansion of the hydroxide and the heat produced during slaking tends to split the material along the particle boundaries. At these break points a new surface capable of further reaction with the pollutants is produced. This is termed the secondary surface increase. The first reaction at the new surface is slaking to form Ca(OH)<sub>2</sub>, followed by the capture of sulphur dioxide.

Hydration or slaking is accomplished by the exothermal reaction



In sulphur dioxide capture, just as in surface increase, a distinction is made between primary and secondary reaction mechanisms.

Starting with calcium hydroxide as the reactant the primary reaction mechanism is as follows:



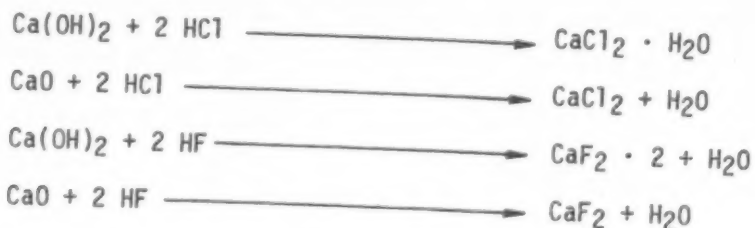
At the fracture zones of the particle the free CaO takes part in the sulphur capture reactions preferentially in the order of the equilibrium constants of these reactions.



These reactions are characteristic for the secondary mechanism.

A comparison of the reaction equilibrium constants shows that for sulfur capture the conversion to calcium sulfate is the preferential reaction.

Simultaneously with the reduction in sulphur dioxide, the HCl and HF contents in the uncleaned gas are reduced. The chemical reactions involved are described by the following equations:



The attainable Desulfurization efficiencies are heavily dependent upon the degree to which saturation of the flue gas is approached. The technical prerequisites for realisation of the flue gas conditioning process are a quench cooler, a flue gas side bypass and (possibly) a flue gas reheat system.

The other process combination for improving calcium utilization, as already stated, is the fly-ash conditioning and recirculation. As the Fig. 1 shows, the principle of fly-ash conditioning is the same as that of flue gas conditioning, namely the reactivation of the free CaO content in the core of the particles covered by  $\text{CaSO}_4$ .

The conditioning process consists in mixing a carefully metered quantity of finely sprayed water homogeneously with the fly ash, separated from flue gas in a filter, in a continuous hydration plant. The water quantity is adjusted to the CaO content of the fly-ash. In a downstream reaction vessel the exothermic chemical reaction then follows.

The fly-ash is treated in such a way as to produce a dry, pourable product which is either added to the fresh additive and injected with it into the boiler, or injected alone. In this way calcium utilisation is improved and a high Desulfurization efficiency achieved for an economically acceptable molar ratio.

The attainable Desulfurization efficiency is heavily dependent on the quantity of recirculated fly-ash and the molar ratio of the Desulfurization process.

The application of direct Desulfurization in combination with quench cooler is recommended especially for boilers equipped with ESP, because flue gas quenching with water helps in upgrading the ESP. Whereas the fly-ash conditioning and recirculation is favourable for boilers with baghouse filter and better quality of by-products. Fly-ash recirculation increases the inlet



mass load in the flue gas highly, which can easily be handled by baghouse filter. Fly-ash conditioning and reinjection in the furnace produces  $\text{CaSO}_4$  and  $\text{Ca(OH)}_2$  as calcium by-products, whereas the method of flue gas quenching  $\text{CaSO}_3$  additionally.

#### 4. DESULFURIZATION RESULTS FROM COMMERCIAL PLANTS

Since the start of work in the field of direct desulfurization in 1980 quite a number of test, demonstration and commercial plants have been installed by Steinmüller. Fig. 2 shows only commercial plants either already constructed and in operation or at the present under construction. As it can be seen, among the boilers in operation there are different types of firing concepts involved e.g. p.f. wall and tangential firing, grate stokers and ignifluid.

In the following the desulfurization results from each type of the above mentioned boilers will be presented.

##### 4.1 Arzberg, FRG

The steam generator of 300 MW thermal capacity has a frontal firing system which has been refitted with a total of 12 SM-burners for low- $\text{NO}_x$  combustion. The 12 burners are arranged in 2 rows of 6 burners each. Limestone injection takes place either through the upper staging air nozzles of the upper burner plane or at the end of the furnace downstream of the furnace constriction. Investigations on the temperature and residence time of flue gases in the furnace show that the lower plane is unsuitable for full-load operation.

Fig. 3 shows the measured desulfurization efficiencies as a function of the molar ratio  $\text{Ca/S}$  for two different additives i.e.  $\text{CaCO}_3$  and  $\text{Ca(OH)}_2$ .

The comparison of results at full load and 60 % load with  $\text{CaCO}_3$  show a considerable increase in desulfurization at 60 % load. The possible reason for the better  $\text{SO}_2$  capture at lower load is of course higher residence time in the proper temperature zone and better dispersion of the additive.

The influence of residence time in case of  $\text{Ca(OH)}_2$  does not seem to be so strong as in case of  $\text{CaCO}_3$ . Hydrated lime is much finer than limestone.

##### 4.2 Malmö, Sweden

The boiler of 125 MW thermal capacity has a tangentially firing system with upper air. The burners are located in the corners. The fuel employed is bituminous coal with a sulfur content of approx. 0.63 %.

Fig. 4 shows the result of desulfurization in full-load and part-load operation. The desulfurization efficiencies have been plotted as a function of molar ratio  $\text{Ca/S}$ . Even for low sulfur coal a reduction of 50 % was reached at molar ratio less than 4 with powdered limestone.

#### 4.3 Salzburg, Austria

In the Salzburg CHP station a direct desulfurization plant was installed in a 30 MW<sub>thermal</sub> boiler operated with reciprocating grate firing system in March 1985. The additive was injected from the front wall via the secondary air nozzles into the furnace.

The desulfurization results are shown in Fig. 5. The ESP behind the boiler is, as discussed later in chapter 5, very small, so that the increase in inlet mass load had to be kept lowest possible. In order to attain the required 50 % desulfurization efficiency, slaked lime was employed instead of powdered limestone because of the better degree of calcium utilisation. The fuel employed was an Austrian brown coal with a sulphur content of approx. 2 %.

#### 4.4 Boras, Sweden

At the end of 1985 the direct desulfurization plant was installed at the Boras CPH station for two spreader stoker boilers each 80 MW thermal capacity. The boilers are operated either with wood chips or imported bituminous coal with low sulfur content of 0.3 to 0.6 %. The desulfurization is in operation only when coal is fired.

Five additive injection nozzles on each side wall of the boiler are located, which are offset with respect to each other.

Fig. 6 represents the sulfur capture as a function of molar ratio Ca/S with limestone at full load and low load of 50 %.

#### 4.5 Schrobenhausen, FRG

At the beginning of 1988 the direct desulfurization plant for ignifluid boiler in Schrobenhausen went in operation. The boiler has a thermal capacity of 50 MW. Two additive injection levels have been selected, lower for partial load and higher for full load.

Fig. 7 shows the sulfur capture as a function of molar ratio. The figure shows the desulfurization efficiency with  $\text{Ca(OH)}_2$  as sorbent at different boiler loads. Due to lack of high capacity demand of the boiler there are till yet no results available at full load and sorbent injection at the upper level.

### 5. OPERATING EXPERIENCE WITH DOWNSTREAM PLANT COMPONENTS

#### Fouling of Heating Surfaces:

Experience so far with boiler fouling during direct desulfurization operation is to be regarded as positive. It is important that the boiler is fitted with sootblowers, as is usual in coal-fired boilers.

It was observed that with direct desulfurization process (DDP) deposits on the heating surfaces and in the flue gas ducts are very loose and light and are easy to clean off.

In some cases it was possible, as a result of the loose nature of the sorbent deposits or their products of reaction, to reduce the number of cleaning intervals. Often it was only in the superheater that these loose deposits were observed.

Only in one case, Boras, was there increased fouling of the economiser as a result of DDP operation. The reason for this was, however, excursions below the dew point in the economiser which resulted from excessive dimensioning of this heating surface after the modification from oil to bituminous coal firing.

No ill effects were observed on the flue gas ducts, ID fan or stack.

#### Electrostatic Precipitator:

No unusual fouling or caking was observed on the electrostatic precipitators during DDP operation.

The prescribed limiting value for dust emissions of 125 to 50  $\text{mg}/\text{m}^3\text{STP}$  was as a rule maintained during DDP operation as well.

Trouble as a result of exceeding the limiting emission values only occurred in those cases where the electrostatic precipitators were underdimensioned even for operation without lime injection, e.g. Salzburg. The electrostatic precipitator at Salzburg consists of only 1 zone with two parallel chambers and is designed for a specific collecting area of 39  $\text{m}^2/\text{m}^3/\text{s}$  (200  $\text{ft}^2/1.000 \text{ acfm}$ ), referred to the projected collecting area. This value is considered extremely low for presentday conditions.

Electrostatic precipitators which have a specific collecting area larger than 75  $\text{m}^2/\text{m}^3/\text{s}$  (384  $\text{ft}^2/1.000 \text{ acfm}$ ), can comply with a limiting value for emissions of 50  $\text{mg}/\text{m}^3\text{STP}$  (0,04  $\text{lb}/10^6\text{BTU}$ ) under normal boiler and fly ash conditions.

In the case of stoker-fired boilers, whose fly ash without lime contains up to 40 % unburnt combustibles, the addition of lime reduces this percentage and has a positive effect upon the precipitation performance. According to the precipitator manufacturers, the optimal content of unburnt combustibles in the fly ash is around 5 - 15 %.

As a result of its specific resistance of only some  $1 \times 10^3 \text{ Ohm} \times \text{cm}$ , a slight proportion of fly carbon in the fly ash tends to improve dust capture. However a higher fly carbon content reduces the performance of the precipitator, because the fly carbon loses its charge very quickly and has only a slight tendency to adhere to the precipitator electrodes. As a result it is easily entrained by the gas flow again.

Since the additive and its reaction products possess however a high specific electrical resistance, this effect is reduced and slight improvement of precipitation efficiency is attained, as long as the critical resistance of approx.  $10^{11} \text{ Ohm} \times \text{cm}$ , which is decisive for reliable electrostatic precipitator performance, is not exceeded.

## 6. DISPOSAL OF BY-PRODUCTS

### 6.1 Introduction

The majority of flue gas cleaning processes produce considerable quantities of by-products. It is extremely important to ensure that these can be disposed of without causing an environmental problem. Thus the safe, long-term disposal of such by-products is an essential requirement for any desulphurisation process, particularly in the light of the legal stipulation that by-products should be utilised as far as possible.

For the flue-gas gypsum from "wet" flue gas processes, procedures have already been developed to render the material suitable for re-use (e.g. drying and compaction). The flue gas gypsum from bituminous coal fired power stations can, in the light of present-day knowledge, be fully utilised in the building and cement industries.

For the flue gas gypsum from brown coal fired power stations intensive development work is currently being carried out to open up new fields of application for this type of gypsum in order to ensure that it can be disposed of in the long term, while complying with the requirements to utilise it as far as possible.

As far as the by-products of direct desulphurisation are concerned, Steinmüller has worked out several possible methods, not merely of disposing of the by-product with the least possible environmental impact but also, in compliance with the requirement for re-utilisation, of rendering it into a useful raw material.

### 6.2 Treatment and Disposal or Re-Utilization

Investigations on the possibility of re-using the by-products from coal firing systems with direct desulphurisation indicate that there are a number of possible means of utilising the material, even though industrial utilisation is currently only practised in a few cases. Nevertheless, for the utilization the pretreatment of the by-product is often necessary.

The treatment of the by-product is usually designed to produce a homogeneous mixture of the DDP by-product and water, plus any additives which may be required.

Here such effects as

- Temperature increase/steam development
- Inclination to caking
- Onset of chemical reactions ("setting")
- Changes in volume

must be taken into account. Any free CaO should be slaked as far as possible.



From a technical point of view these problems have largely been solved. Normally two-stage processes are employed, which ensure complete hydration of the free lime.

Because of the well-known difficulties in utilising the by-products from DDP (e.g. as a result of variations in the composition) for safety reasons the possibility of dumping the material must be provided in every case.

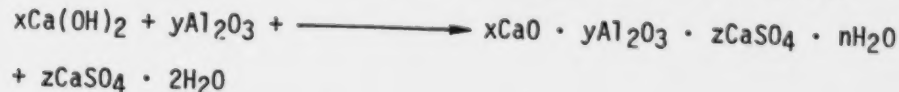
However the following possible methods of utilisation are also given:

- Cement, concrete and building material industries
- Landscaping
- Constructing roads and tracks
- Neutralisation of sludges, e.g. from production processes
- Use as a filling material in deep mines

When mixed with water the calcium hydroxide reacts in a first hardening phase mainly with the silicon dioxide, aluminium oxide and iron oxide constituents in the fly ash, in accordance with the reactions



and, in a second hardening phase in the presence of calcium sulphate, with this and the aluminium oxide in accordance with the reaction



thus producing a material with a high degree of mechanical strength, low permeability and low elution values.

Since the commencement of development work on direct desulphurisation Steinmüller has already explored the disposal problems connected with its use.

One aspect of this is the intensive contacts and cooperation with disposal companies.

Among the various attempts can be listed marketing of the material as a means of filling up old mine workings and, in the longer term, as an auxiliary material for the building industry. As far as use as a mine working filler is concerned, treatment plants with a capacity of several hundred thousand tonnes per year are currently being installed.

After suitable treatment the fly ash is then used to prevent damage resulting from mining subsidence, in particular by compacting the slopes of mine tailings and filling up old workings.

Such landfill using power station by-products has in some cases already been licensed by the competent Federal German authorities. Further licences have been applied for and the competent licensing authorities have already shown a positive reaction to the proposed measures.

There are also concrete possibilities in the Federal Republic of Germany for further applications as an additive in road making and in the construction of railway embankments, such as is already practised in Sweden and licensed by the authorities there.

As is well-known, lime is used for drying sewage sludge. Because of the relatively high calcium content of DDP fly ash, this by-product is also suitable for drying sewage sludge and is currently being used in a number of sewage farms.

The position is thus that there are already a number of firms who specialise in the disposal of fly ash from direct desulphurisation plants. These firms are also prepared to conclude long-term disposal agreements.

### 6.3 Fly Ash Disposal in Some Power Stations

#### Malmö

In 1984/1985 the municipal district heating power station in Malmö concluded a disposal agreement with Messrs. CEMENTA who carried out substantial tests and investigations concerning the use of DDP fly ash in cement manufacturing.

The main objections raised by the authorities concerned both reactivity and the pozzolana factor which render the use of DDP fly ash in the cement industry difficult. The problem is particularly acute with those coals which produce very high furnace temperatures, since under these conditions the free CaO particles tend to sinter. This in turn leads to a very slow reaction for the conversion to  $\text{Ca(OH)}_2$  and prevents the material from setting properly. In order to offset this disadvantage, CEMENTA has developed a new process which is marketed under the name CEFYCL and allows the use of DDP fly ash in cement manufacturing and road construction.

#### Salzburg

In the Salzburg combined heat and power station, stoker bottom ash and DDP fly ash were originally stored in a common bunker where they were blended. This solution, however, could not be continued as it led to caking. As a consequence, a separate fly ash bin was installed and the bottom ash, which was thus no longer contaminated by the injected limestone, could be sold for use in the building material and cement industry.

The DDP fly ash was transported in a dry or wet condition to a hazardous waste dump.

The requirements imposed by the Austrian authorities prevented the adoption of any other solution.

At present, investigations are being carried out in order to find further applications for DDP fly ash in the foreseeable future.

#### Arzberg

Without DDP, the fly ash from this brown coal power station makes a first-class filler material for cement manufacturing.

With the direct desulfurization plant in operation, however, the fly ash is no longer suitable for this purpose. Part of the DDP fly ash was therefore used for clinker manufacturing and the rest was disposed of by dumping.

Tests performed in order to find out whether the DDP fly ash is suitable for dumping showed that the material can be used for dumping in old excavations.

The geotechnical report raised no objections against proper disposal of the DDP fly ash in dumping sites, either with regard to its loadcarrying capacity or the risk of contamination of soil or ground water.

Permission for dumping was made conditional on lining the bottom of the excavation with a compacted layer of DDP fly ash.

The DDP fly ash was tipped after the free CaO had been slaked (max. 15 %) and sufficient water for setting had been added. In this way a concrete-like mass was obtained (pozzolanic reaction).

Examinations of trepanned cores consisting of unpacked material produced the following mean values:

|                              |                    |                   |
|------------------------------|--------------------|-------------------|
| 28-day compressive strength: | 11                 | MN/m <sup>2</sup> |
| Density in moist condition:  | 1.5                | t/m <sup>3</sup>  |
| Permeability:                | 6-10 <sup>-8</sup> | m/s               |

Leaching, e.g. of sulfates, was only observed round the edges and then only with continuously flowing water.

#### Merck, Darmstadt

Messrs. Merck concluded a DDP fly ash disposal agreement with Messrs. Rheinbraun.

Under this agreement, the by-products can be disposed of if required in opencast mining excavations of the Rhenish brown coal field.

At present, the DDP fly ash is not disposed of by dumping but it is sold to a processing company. We do not have any further information on the company or the application selected for the DDP fly ash.

Schrobenhausen, Leinfelder paper mill

The DDP fly ash from the ignifluid firing system is utilised by the cement industry for clinker manufacturing.

The fly ash serves to grog the rich clay which is used in manufacturing the klinker.

This method of fly ash disposal is not new, having already been practiced for more than 20 years (Arzberg, Schwandorf).

DISCLAIMER

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.



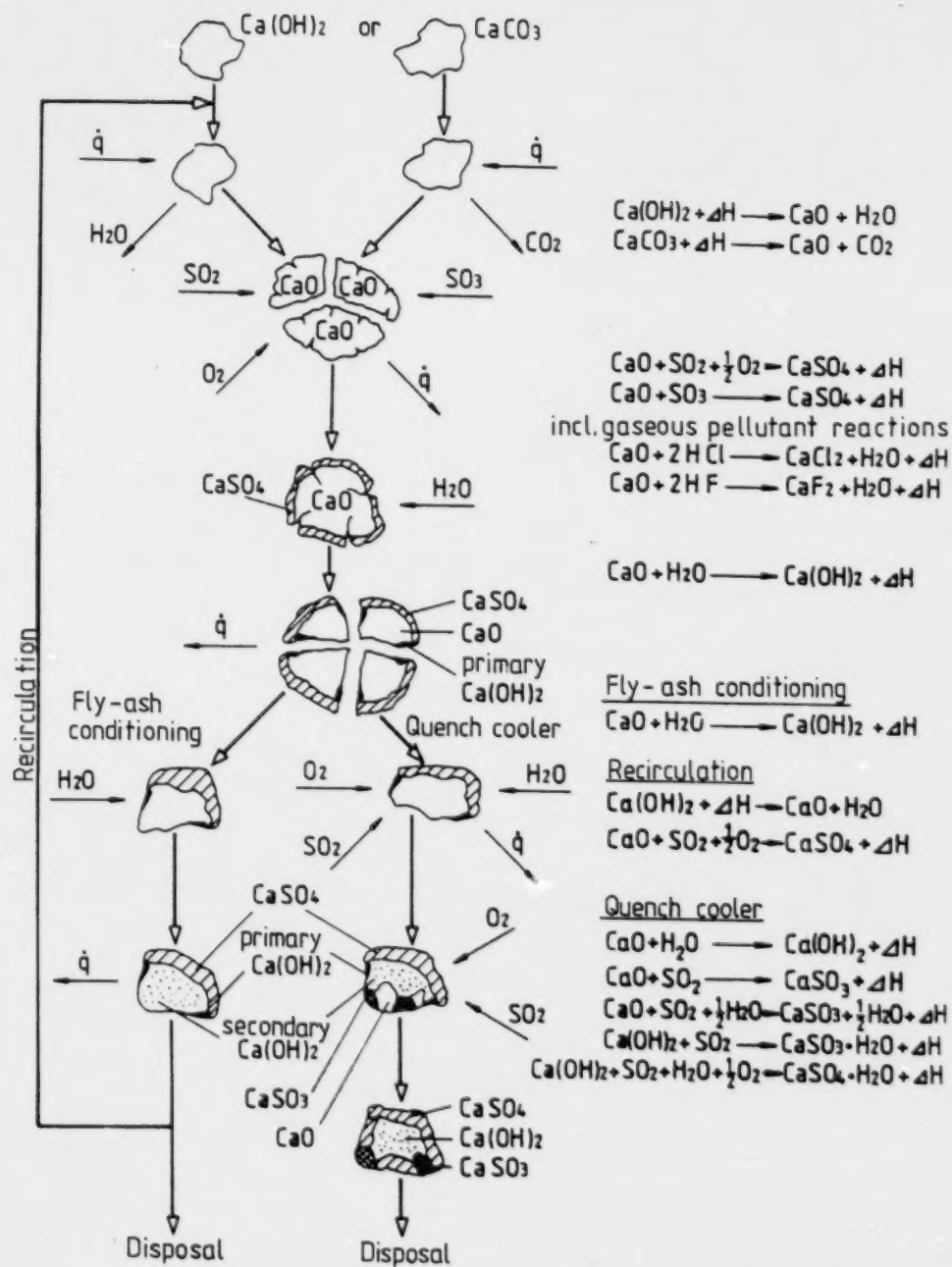


Fig. 1: Schematic representation of Direct Desulfurization Process

|                          |     |                          |                     |      |
|--------------------------|-----|--------------------------|---------------------|------|
| 5. CFB Gagetown          | CDN | Circumferential burner   | 2 x 12MW            | 1984 |
| 6. Merck Darmstadt       | D   | Travelling grate stoker  | 92MW                | 1984 |
| 9. Borås Kraftwerk       | S   | Travelling grate         | 2 x 80MW            | 1985 |
| 12. Arzberg              | D   | Frontal firing system    | 300MW               | 1984 |
| 13. Malmö                | S   | Tangential firing system | 125 MW              | 1985 |
| 14. Salzburg             | A   | Stoker firing system     | 30MW <sub>el</sub>  | 1985 |
| 18. Schrobenhausen       | D   | Ignifluid firing system  | 49MW                | 1987 |
| 19. Paris                | F   | Ignifluid firing system  | 2 x 350MW           | 1989 |
| 21. New Brunswick Power  | CDN | Tangential firing system | 107MW <sub>el</sub> | 1988 |
| 22. Boehringer Ingelheim | D   | Travelling grate         | 65MW                | 1989 |
| 23. Puente Nuevo         | E   | P.f. shoulder firing     | 140 MW              | 1989 |
| 24. Berlin - Neukölln    | D   | Travelling grate         | 3 x 27MW            | 1989 |

Fig. 2: List of commercial plants either in operation or in design phase

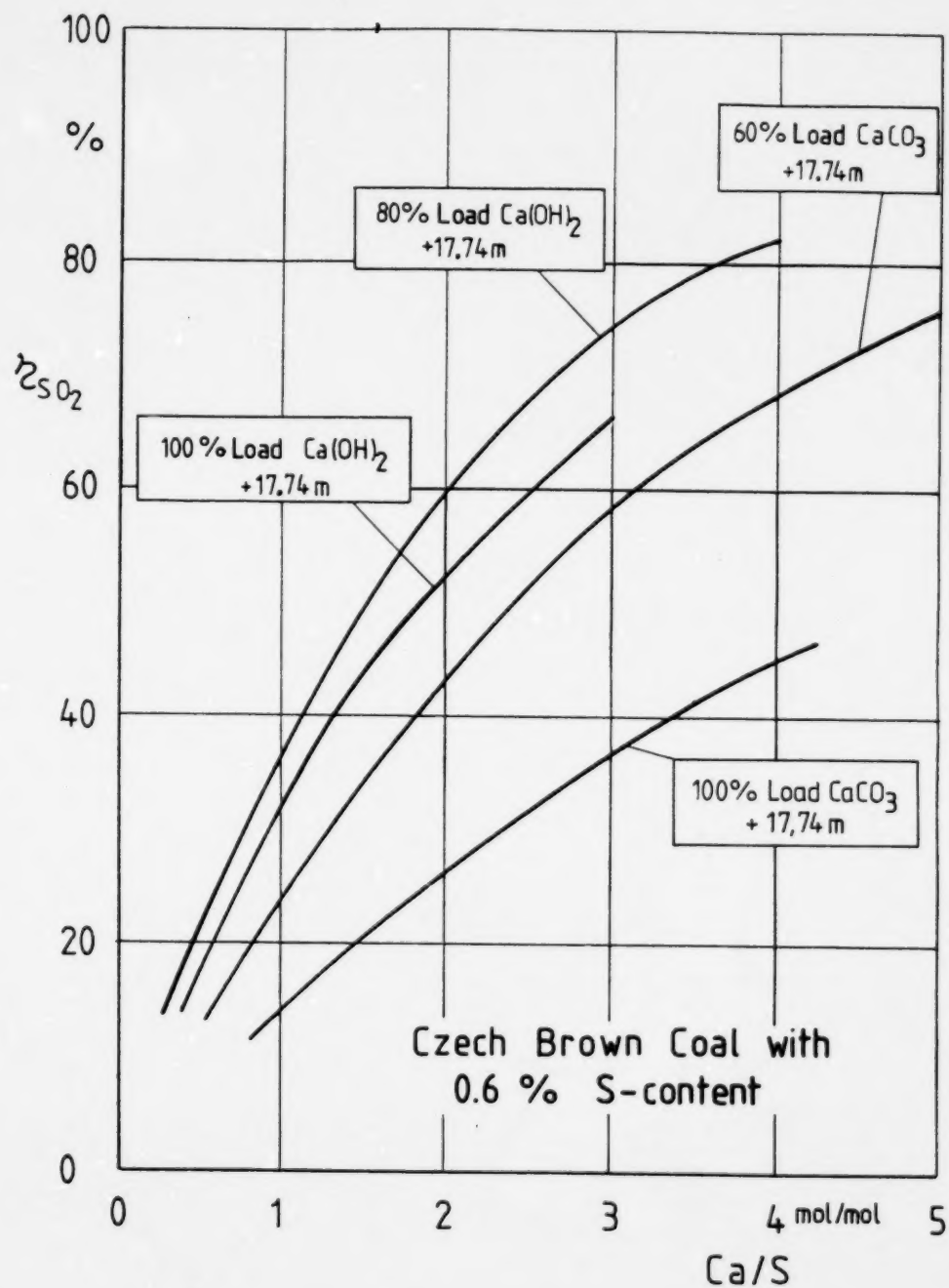


Fig. 3: Desulfurization efficiency as a function of molar ratio  $Ca/S$  at Arzberg, W. Germany

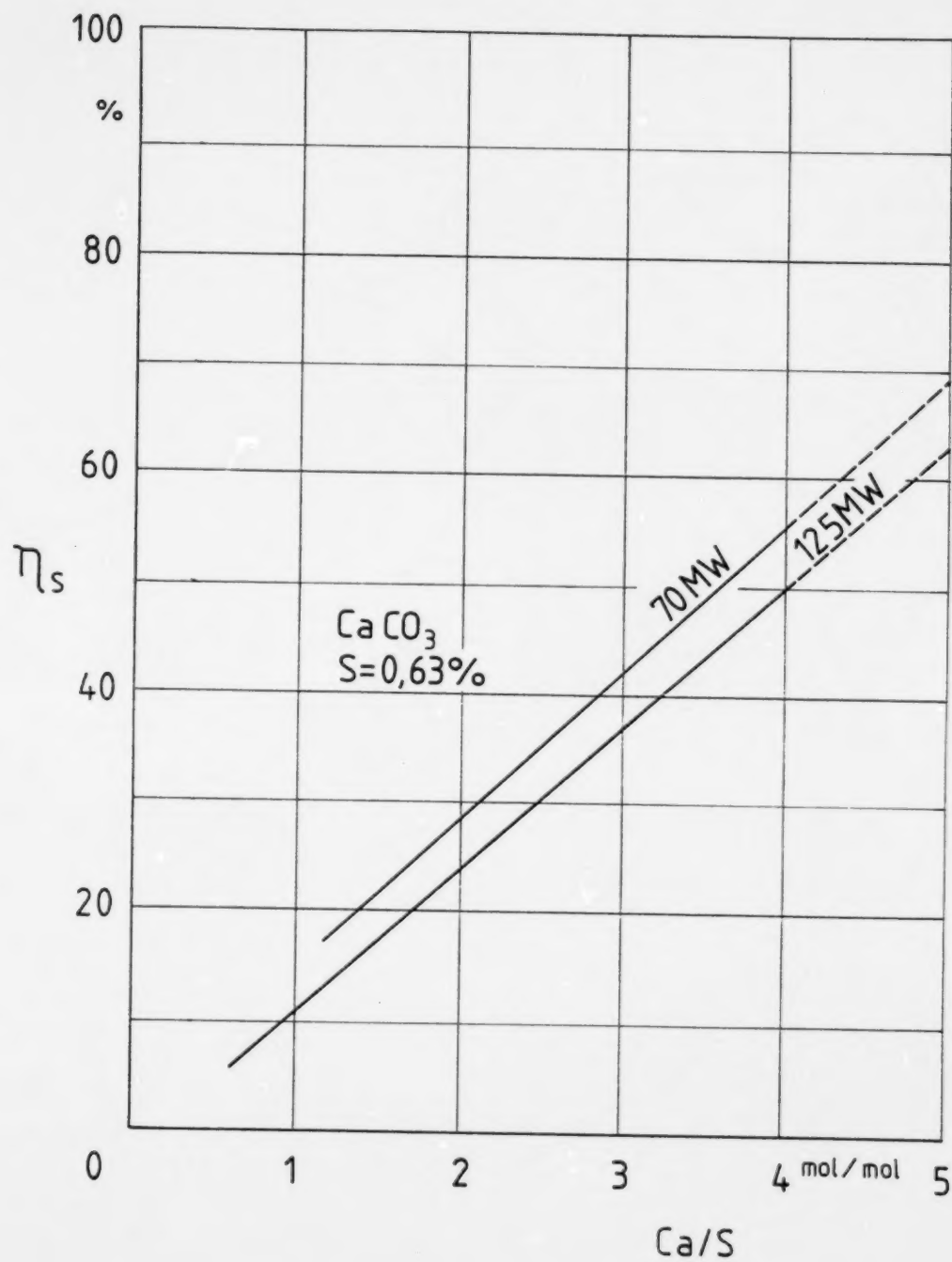


Fig. 4: Desulfurization efficiency as a function of molar ratio  $\text{Ca/s}$  at Malmö, Schweden



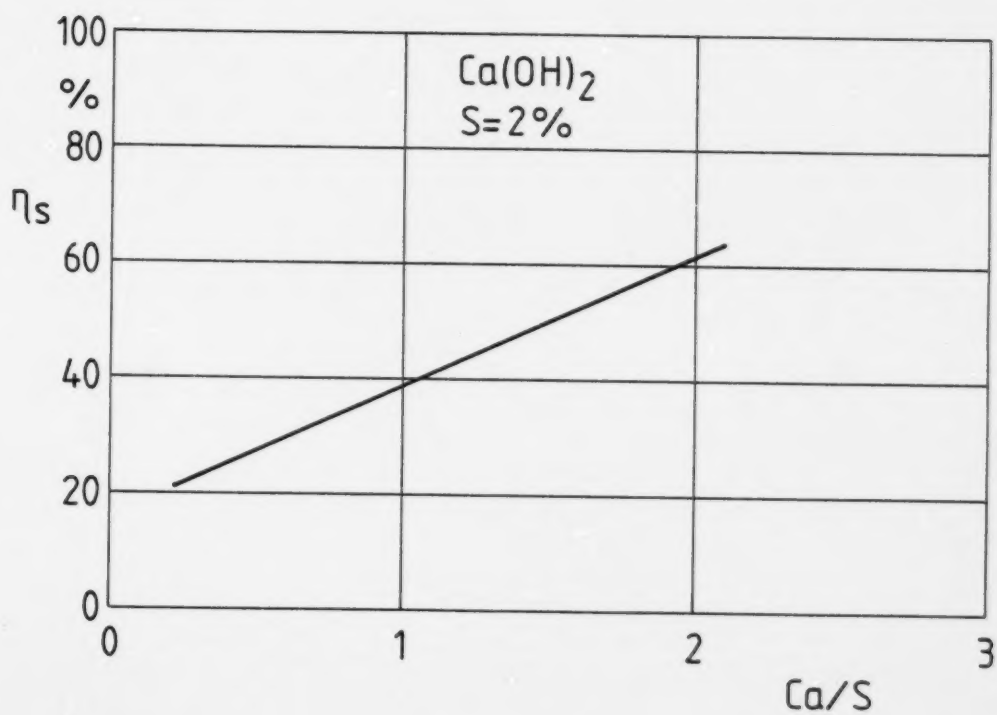


Fig. 5: Desulfurization efficiency as a function of molar ratio  $\text{Ca}/\text{s}$  at Salzburg, Austria

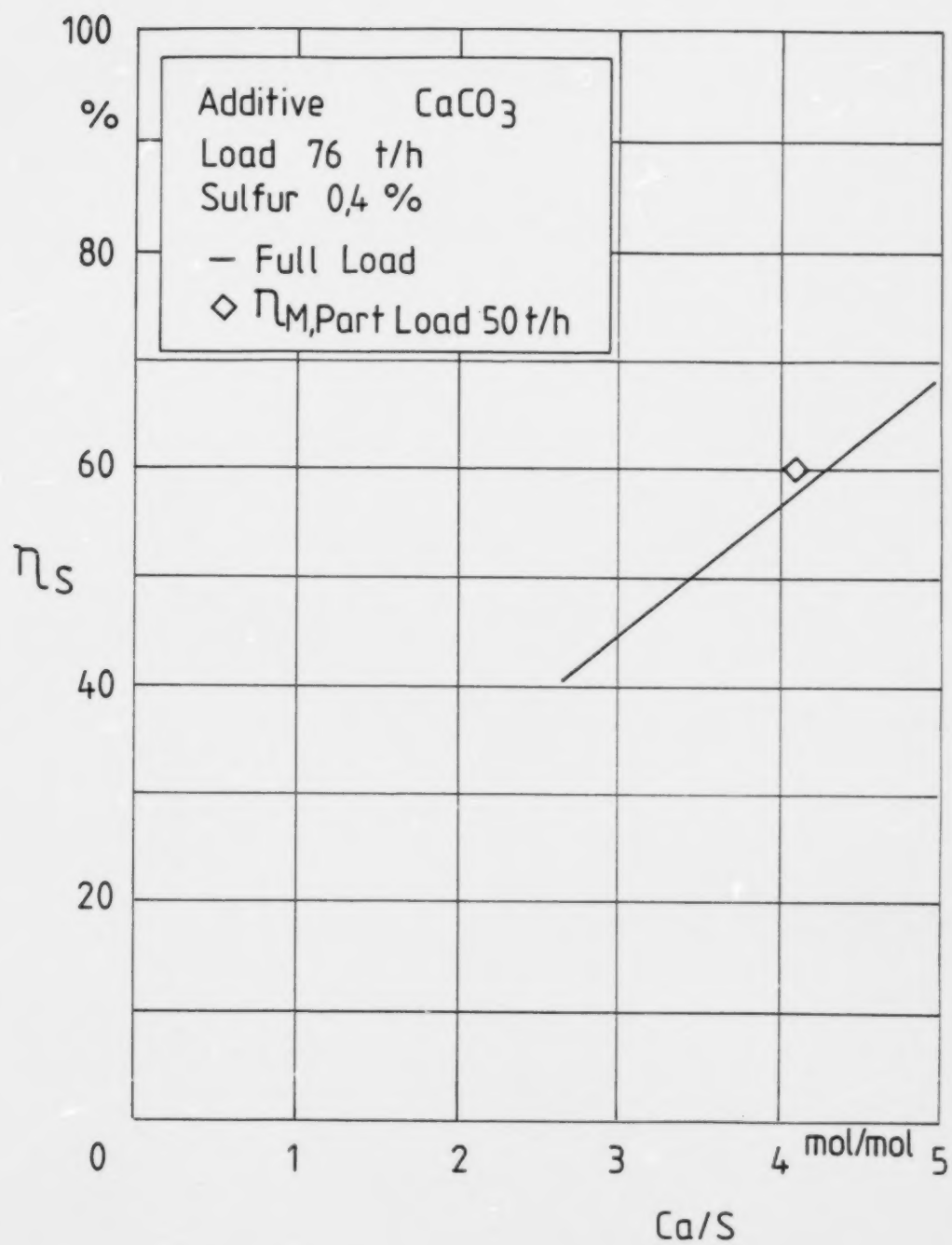


Fig. 6: Desulfurization efficiency as a function of molar ratio Ca/s at Boras, Sweden

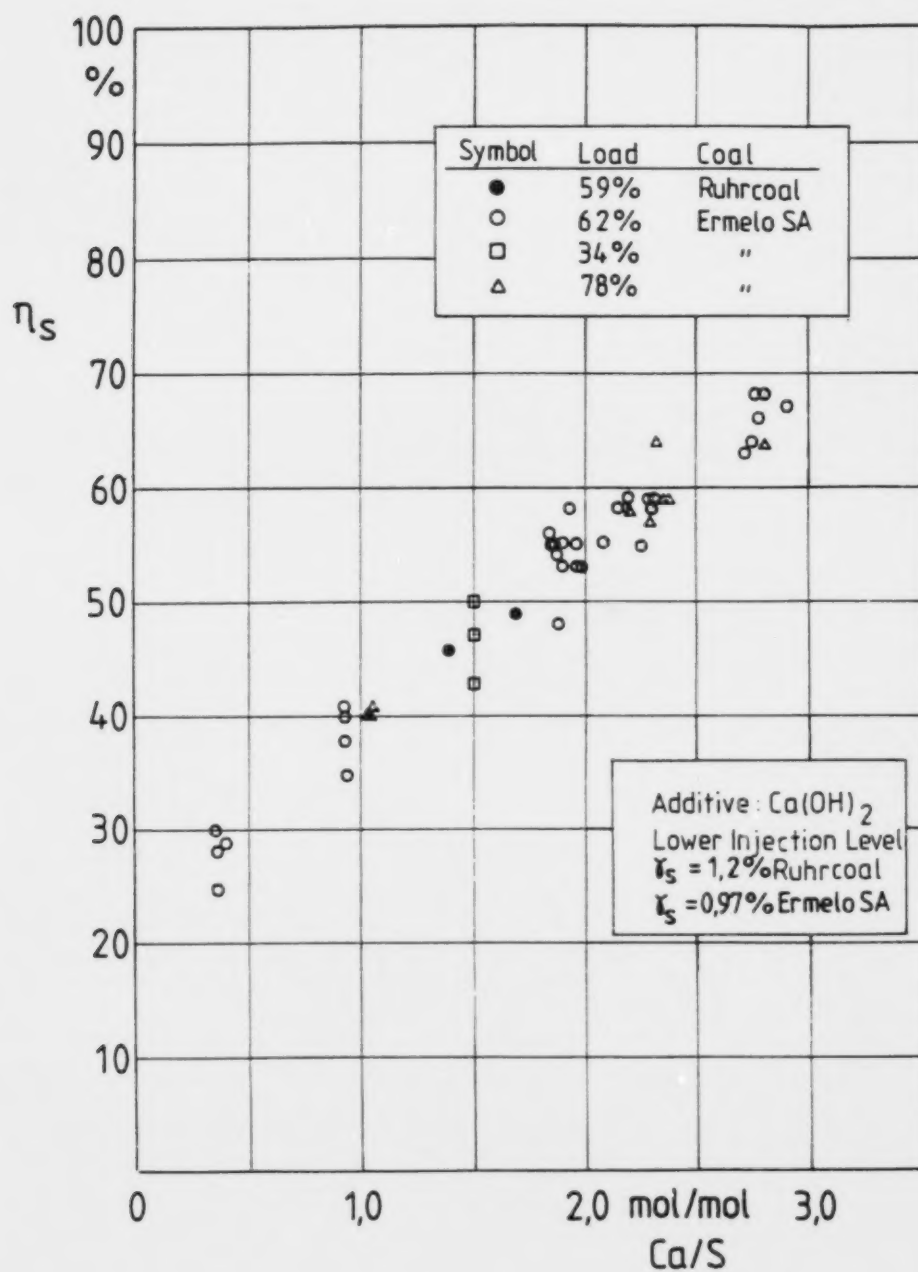


Fig. 7: Desulfurization efficiency as a function of molar ratio Ca/s at Schrobenhausen, W. Germany

## AUSTRIAN EXPERIENCE WITH FURNACE LIMESTONE INJECTION

by: G. Staudinger, P. Melcher  
Graz University of Technology  
A 8010 Graz, Austria

K. Eckerstorfer  
Oberösterreichische Kraftwerke AG (OKA)  
Böhmerwaldstraße 3  
A 4020 Linz, Austria

### ABSTRACT

In Austria, twelve boilers are equipped with furnace limestone injection (FLI), eight of them still use FLI in order to comply with the law. Until now they have accumulated more than 125 000 hours of operational experience. The limits set by the new law regulating the emission levels are such that for most plants of less than 150 MWth capacity the utilization of low sulfur coal in combination with FLI plus some improvement like duct injection of activated, recycled ash (ARA) will be sufficient to enable them to comply.

The ARA process has been further developed as an addition to FLI. It has been proven that it can achieve (in combination with FLI) 80% sulfur removal without a baghouse and 90% removal if a baghouse is used. Some additional heat can be recovered from the flue gas without confronting us with corrosion problems.

A new method of FLI ash dumping yields an impermeable body with high compression strength. FLI ash can now be disposed of in an environmentally safe way.

### NEW EMISSION STANDARDS FOR AUSTRIA

In summer 1988 a law was passed by the Austrian parliament which introduced new emission standards for both old and newly built plants. Figure 1 represents the emission standards for solid fuels applying to plants of different size in Austria in comparison to the emission limits of the two neighbouring countries Germany and Switzerland, which restrict sulfur emission. The limits for plants which burn brown coal are slightly higher than for those which burn other solid fuels.

On the basis of this law the utilities can and must now decide what they will do with the existing plants. They have to submit their proposals to the authorities by Jan. 1st 1990. If the authorities approve the proposals, these must be implemented within three years.



For Timelkam No 2 (see table 1) e.g. it became clear that with its capacity of 170 MWth the emission limits for  $\text{SO}_2$  will be  $600 \text{ mg/Nm}^3$ , which cannot be guaranteed by furnace limestone injection, but can easily be obtained by an extended furnace limestone injection process, i.e. the ARA-process, which shall be described in a later chapter. Riedersbach No 1 has less than 150 MWth; it will probably be approved if it changes to burning low sulfur fuel in combination with FLI.

#### STATUS OF FURNACE LIMESTONE INJECTION

In Austria, twelve boilers are equipped with FLI as shown in table 1. Nine of them are still in operation using FLI. Three plants no longer use FLI but they still have the installation as a backup.

Voitsberg No 2 has had very few operating hours since 1984 and had been conserved as an emergency capacity in 1987. The law allows operation of another 5 000 hours of operation with FLI before it is dismantled.

St.Andrä No 2 is complying with regulations introduced in 1985, i.e. is an 80% sulfur removal level, irrespective of the  $\text{SO}_2$  concentration. When it was first imposed this level could not be guaranteed by a dry process. ÜDK developed as an addition to FLI, the sodiumb carbonate process (1, 2).

Voitsberg No 3 installed FLI as an intermediate solution until the completion of the wet FGD which has been treating the full flue gas flow since 1986. The FLI is still there and operable for cutting peaks of extremely high sulfur fuel in case the FGD should not be able to keep the emission limit. This however has not been necessary until now. Operating data on Voitsberg No 3 were published earlier (1, 2, 3). There was no major problem caused by FLI.

The cogeneration plant of Graz has three identical boilers designed for burning of both brown coal and oil. In order to use the offgas coming from a nearby waste disposalsite one of the boilers was fitted with additional equipment for burning gas. It has been operated on gas only since 1987.

Riedersbach No 1 and Timelkam No 2 have been run on a variety of brown coals with FLI, causing no difficulties worth mentioning. An experiment on ash disposal is described later.

Riedersbach No 2 had FLI, by utilizing the roller mills - which are provided for hard coal grinding - to prepare limestone powder, only as a backup until the wet FGD plant was in full operation. Today Riedersbach No 2 no longer uses FLI.

The cogeneration plant of Klagenfurt has four boilers, three of them can burn coal. Since the installation of FLI there have been no problems as regards the flue gas side of the three boilers. To the knowledge of the author this is the only plant where the powder flow to the furnace is controlled automatically on the basis of the  $\text{SO}_2$  concentration in the stack. The system worked well until the star valve failed. The system will be operable again in the fall, when it is required. The boilers of the cogeneration plant will be in compliance with the new law, if they burn low sulfur coal in combination with FLI.

In short it can be said that this paper contains little news compared to the last three papers (1, 2, 3). All plants have been operating well, meeting the sulfur removal targets. The new plants were equipped with a FGD for the full flue gas flow. The older and smaller plants will continue to run on FLI, some of them with an additional dry sulfur removal stage in order to meet the emission limits.

## EXPERIENCE IN OROSZLANY (HUNGARY)

One FLI plant was sold to a power station in Hungary with 55 MWe1 capacity. The fuel is brown coal the analysis of which is given in table 2. With the very high SO<sub>2</sub> concentration of 5 000 to 6 000 ppm in the flue gas very good utilization of the added limestone is achieved as the ash analysis in table 3 shows.

For better limestone utilization the ARA process is envisaged for future applications.

## NEW DEVELOPMENTS

### LIMESTONE INJECTION AND SUPPLEMENTAL SODIUMBICARBONATE

In earlier papers ÜDK described the development of a process which injects sodiumbicarbonate (NaHCO<sub>3</sub>) into the flue gas duct between the air preheater and the fly ash precipitator (2). In a later paper (3) the installation of a full scale unit in St.Andrä was announced.

The combination of furnace limestone injection and duct injection of sodiumbicarbonate was chosen and is still in operation for economic reasons: NaHCO<sub>3</sub> is very expensive and not available on the market in high quantities and at short notice, and thus requires stocking. Therefore FLI is used to eliminate about 50% of the sulfur, the rest is absorbed by the sodiumbicarbonate. Clean gas concentrations of SO<sub>2</sub> as low as 200 mg/Nm<sup>3</sup> can be obtained, irrespective of what the raw gas concentration is. Research work at St.Andrä No 2 focuses on an improvement of the FLI in order to save sodiumbicarbonate without adding more limestone.

### IMPROVEMENTS IN FURNACE LIMESTONE INJECTION

There is an incentive to improve the efficiency of FLI in order to

- o save supplemental sodiumbicarbonate
- o save limestone powder
- o avoid installation of more expensive technology to meet emission requirements.

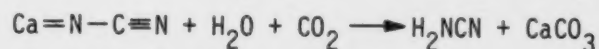
### Method of Injection

During a boiler shut-down in summer of 1988 the burner arrangement of St.Andrä No 2 was modified mainly with the aim of reducing NO<sub>x</sub>. In a test run a dramatic improvement of sulfur removal from 35% to 64 and 62%<sup>x</sup> with the same limestone powder at the same Ca/S ratio of approximately 4.4 was observed at full boiler load. St.Andrä No 2 was known to give the poorest limestone utilization of all Austrian plants. With this alteration in burner geometry it was shown that there is still some potential for improvement in FLI.

### Special Limestone Powder from SKW

SKW (Süddeutsche Kalkstickstoffwerke) produces fertilizers and chemicals on the basis of calciumcarbide CaC<sub>2</sub>; one of these being calciumcyanamide.

SKW-limestone is a calciumcarbonate coming as a by-product from the reaction of calciumcyanamide with water and CO<sub>2</sub> in the production of cyanamid.



The calciumcarbonate is contaminated with Ca(OH)<sub>2</sub> and elemental carbon. Until now it was considered as a waste. Experiments in St.Andrä No 2 and Riedersbach No1

showed that this fine, precipitated material can replace natural limestone powder with at least equal utilization of the calcium.

In the spring of 1988 (thus before the burner arrangement was changed) ØDK conducted a number of well documented tests with SKW-limestone with the following results:

#### St.Andrä No 2, ØDK

| load, MWel | limestone | Ca/S | SO <sub>2</sub> -removal % |
|------------|-----------|------|----------------------------|
| 80         | ØDK       | 4.5  | 45 - 50                    |
|            | SKW wet   | 3.4  | 56                         |
|            | SKW dry   | 4.2  | 78                         |
| 100        | ØDK       | 4.8  | 35 - 40                    |
|            | SKW wet   | 3.4  | 40                         |
|            | SKW dry   | 3.8  | 56                         |

"SKW wet" means that the limestone was added in the wet state, as it was produced as filter cake with a water content of 20 - 24% to the coal and processed with the brown coal through the beater mills. "SKW dry" is the same material, but dried in a rotating drum dryer and injected into the furnace in a conventional way.

The improvement can easily be seen from the figures: At lower stoichiometric ratios the SO<sub>2</sub> removal rates are markedly higher. The somewhat poorer results with the wet material are not surprising, since it has been known for a long time that injection with overfire air is a more effective method than addition to the coal (4).

#### DEVELOPMENT OF NEW DUMPING METHOD FOR FLI ASH

In a paper published by ØDK in 1985 (2) Schröfelbauer et al showed (figure 2) that the composition of ash obtained from brown coal is changed in the direction of cement if it is enriched with CaO through FLI. Although there is little chance that clinker will form - and no additional grinding - a greater tendency to form concrete-like aggregates than with normal ash is to be expected. Brief trials to use it for the manufacture of building bricks showed that this is possible in principle, however there is little need for it on the market.

Riedersbach No 2, in 1987, received permission for a large-scale dumping experiment with FLI ash. There is space for 100 000 tons of ash. From October 1987 to April 1988 30 000 tons of FLI ash were dumped following a method which was worked out in the laboratory of OKA.

The tests and measurements conducted in connection with this experiment showed excellent results:

|                       |                       |                         |
|-----------------------|-----------------------|-------------------------|
| Permeability:         | K-value after 28 days | $2.2 \cdot 10^{-8}$ m/s |
| Compression strength: | after 28 days         | 19.2 N/mm <sup>2</sup>  |

An elution test, accomplished with deionized water and 5 bar differential pressure on a block of solidified ash yielded a liquid, the heavy metals content of which was below the limits for drinking water, except for chromium, which was somewhat higher, but still better than the standard of dump sites for building materials.



Having this dumping technology available FLI ash can be disposed of in an environmentally safe manner. Moreover, the properties of the ash allow its use as a sealing material for other disposal sites.

#### ACTIVATION OF RECYCLE ASH (ARA)

Furnace limestone injection is an effective and low-cost method to control sulfur dioxide emissions from coal fired boilers. It has, however, two drawbacks, i.e. the poor utilization of 10 - 15% of the additive (2) and insufficient sulfur removal rates of only 50% at reasonable Ca/S ratios. Most authorities impose higher sulfur removal rates and an emission limit value which can not be brought about by FLI. For this reason FLI was combined with the sodium bicarbonate process in St. Andrä.

Another way to achieve better sulfur dioxide removal in conjunction with FLI is to utilize the CaO, which is present in the FLI ash. For this an attempt was made to increase the residence time of the CaO in the flue gas by means of a baghouse and at the same time to saturate the flue gas with water. In order to get effective absorption of SO<sub>2</sub> in the baghouse the flue gas temperature must be lowered to a level very close to the dew point.

The method of ARA, i.e. removing FLI ash from the flue gas, activation of the CaO with steam and reinjection of that ash into the flue gas duct upstream of the dust precipitator according to figure 3, was first published in (4). Results obtained with a pilot plant on a 500 m<sup>3</sup>/h scale (3) showed that this process allows an effective SO<sub>2</sub>-removal at rates of up to 80 and 90%, with the Ca/S ratios which give only 50% removal in FLI.

#### Pilot Plant Tests at Timelkam No 2

After the promising results achieved with a pilot plant at the cogeneration plant of Graz (3) a somewhat larger pilot plant was set up at Timelkam No 2 in order to test the process with a different ash and to evaluate design data for a full scale unit. Further it was to be investigated whether the baghouse is absolutely necessary for sulfur removal or whether an ESP could also be used as dust collector.

The pilot plant was set up as illustrated in figure 4. Flue gas was taken from the duct downstream of the ESP and cooled by convective heat exchange to a temperature T<sub>1</sub>. After addition of the treated ash, the flue gas went either directly to the baghouse or it passed through a cyclone in which most of the fly ash was collected. From the baghouse the flue gas was released into the atmosphere.

Fly ash was taken from the ESP of the power plant and brought into the steaming vessel through a lock hopper. An adjustable screw feeder supplied a continuous flow of treated ash to the experimental flue gas duct. After its first term the ash was repeatedly recycled to the steaming vessel until the end of the experiment. The activity of the ash increased during the first two or three cycles, then it remained constant for at least ten cycles.

Table 4 shows some of the results obtained with the pilot plant in the cyclone version (2 in figure 4). In these tests the flue gas flow was 850 to 900 m<sup>3</sup>/h, the flue gas temperature T<sub>1</sub> was 74 to 75°C. In table 4, CABE stands for the calcium load in the flue gas flow after the addition of the treated ash. SO<sub>2</sub>IN is the SO<sub>2</sub> concentration at the flue gas cooler, i.e. before addition of treated ash. CA/S is the atomic ratio of the total calcium in the ash to the sulfur in the gas and ESGRD means the removal percentage of SO<sub>2</sub> from the gas phase.



As the results in table 4 show, there is a direct relation between the Ca/S ratio and the sulfur removal. The baghouse is not essential for the functioning of the process. However, as we know from earlier results, a baghouse would be about 20 percentage points more efficient than the cyclone alone.

#### Conception of an ARA for Timelkam No 2

With a rating of 170 MWth and brown coal as fuel, Timelkam No 2 will have to meet an emission limit of 600 mg/Nm<sup>3</sup> from presumably 1994 onwards. The low sulfur fuel which is used in Timelkam No 2 leads to a SO<sub>2</sub> concentration of 2 000 to 2 500 mg/Nm<sup>3</sup>; a reduction to 600 mg/Nm<sup>3</sup> is obtained with 76% sulfur removal in the worst case. The ARA-process is able to achieve this in its simple version with the cyclone arrangement, but without a baghouse, because 50% SO<sub>2</sub> removal is got from FLI and 60% is achievable with a cyclone, thus  $50\% + 0.5 \cdot 60\% = 80\%$ . The existing ESP may be used as dust precipitator. In Timelkam No 2, the existing ESP is a newly built retrofit unit, which was erected adjacent to the boiler house as shown in figure 5. There is a rail track between the boilerhouse and the ESP, and there are other rails behind the ESP. The flue gas comes from two parallel air preheaters and is kept as two separate flows as far as and including the two i.d. fans.

In the situation of Timelkam No 2 any retrofit installation should:

- o use the ESP
- o take care of the rails and other nearby installations

The given situation leads to a setup for the ARA for Timelkam No 2 as shown in figure 6. The flow direction in the ESP is inverted. As a result each of the two flue gas flows is passed by the ESP to two pairs of cyclones, giving a sufficiently long, straight piece of channel which allows effective and homogeneous humidification of the flue gas. The two cyclone exit-flows are united before they enter the one half of the ESP. Downstream of the air preheater, some of the heat will be recovered by convective heat exchange for the purpose of district heating. The amount of heat recovered depends on the requirements of the district heating system. The rest of the cooling is accomplished by injection of water. The treated ash is injected upstream of the cyclones from at least two opposite nozzels. The ash will be collected from both ESP and cyclones, and treated in the steaming vessel. Transportation of the ash is pneumatic by steam.

#### Test program for 1988/1989

In order to provide design data and to get some experience in the operation of the individual components the following test program will be carried out:

- o Test of water spraying in one flue gas duct for the
  - selection of nozzles
  - optimization of nozzle arrangement
  - exploration of lowest possible flue gas temperature
  - function of ESP with respect to collection efficiency and ash handling
- o Further tests with the existing pilot plant in order to obtain more process design data.
- o Tests in a new pilot plant with an experimental ESP.

The test results enable OKA to design one half of an ARA plant for half the flue gas flow for Timelkam No 2. The full plant must presumably be operable before 1994.

## Costs

The cost of an ARA plant is estimated to be one third to one half of an equivalent wet scrubber system.

## ACKNOWLEDGEMENTS

Fundamental research into furnace limestone injection was supported by the Fonds zur Förderung der wissenschaftlichen Forschung. The ARA research and development project was supported by the Austrian Ministry of Science and Research, with Dipl.Ing. Zellhofer as the project officer.

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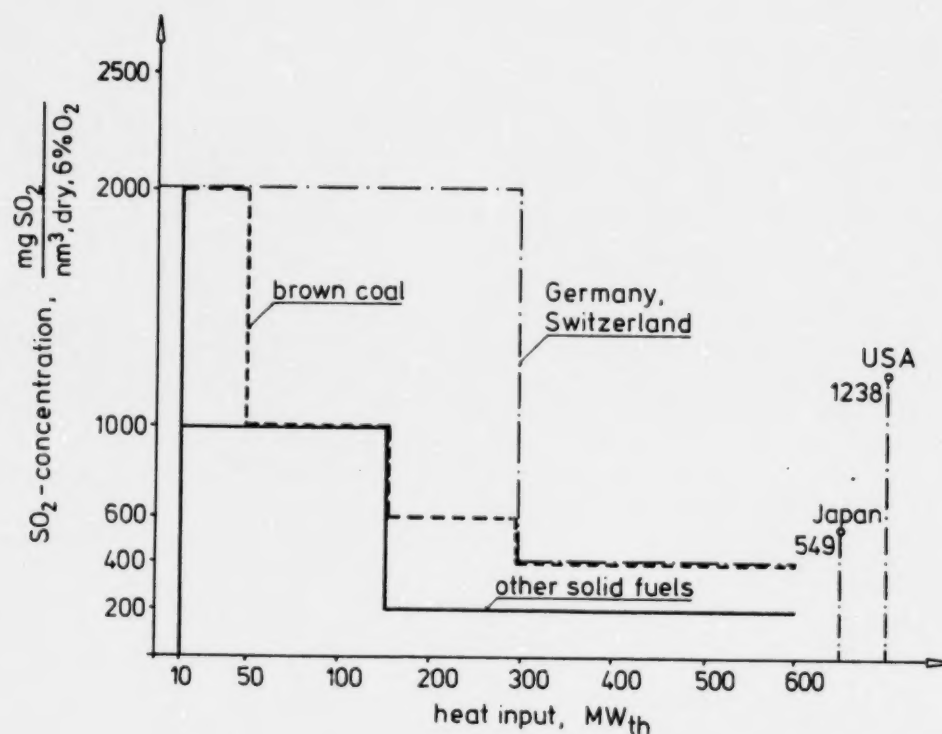


Figure 1. Emission standards in Austria as of Jan 1st 1989.

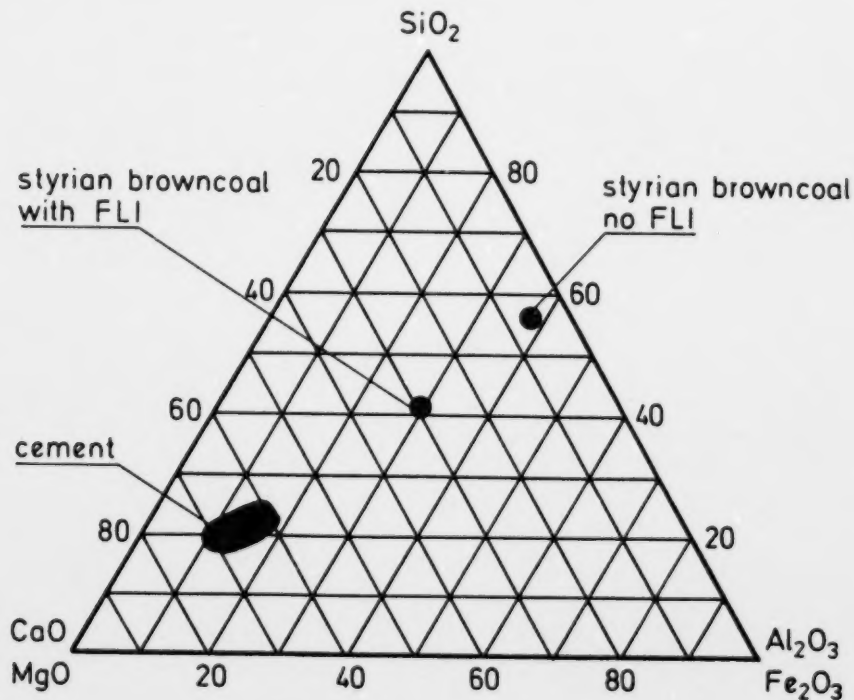


Figure 2. Diagram showing mayor components of cement and fly ash from styrian brown coals (with courtesy of ÖDK).

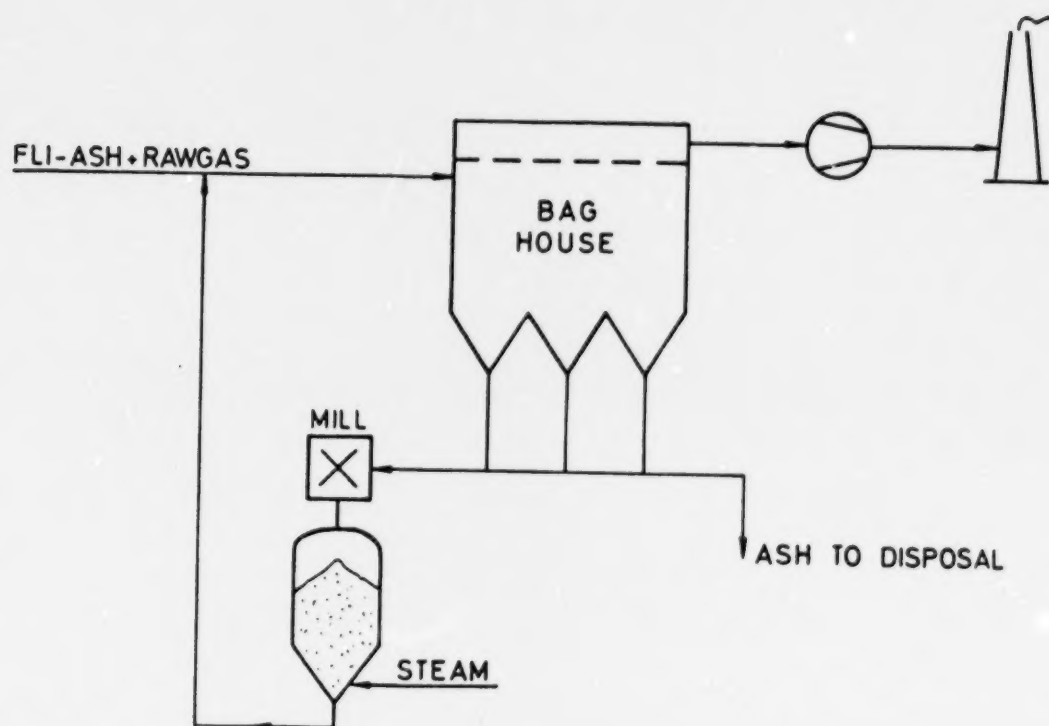


Figure 3. The Ash Recycle Activation Process (ARA).

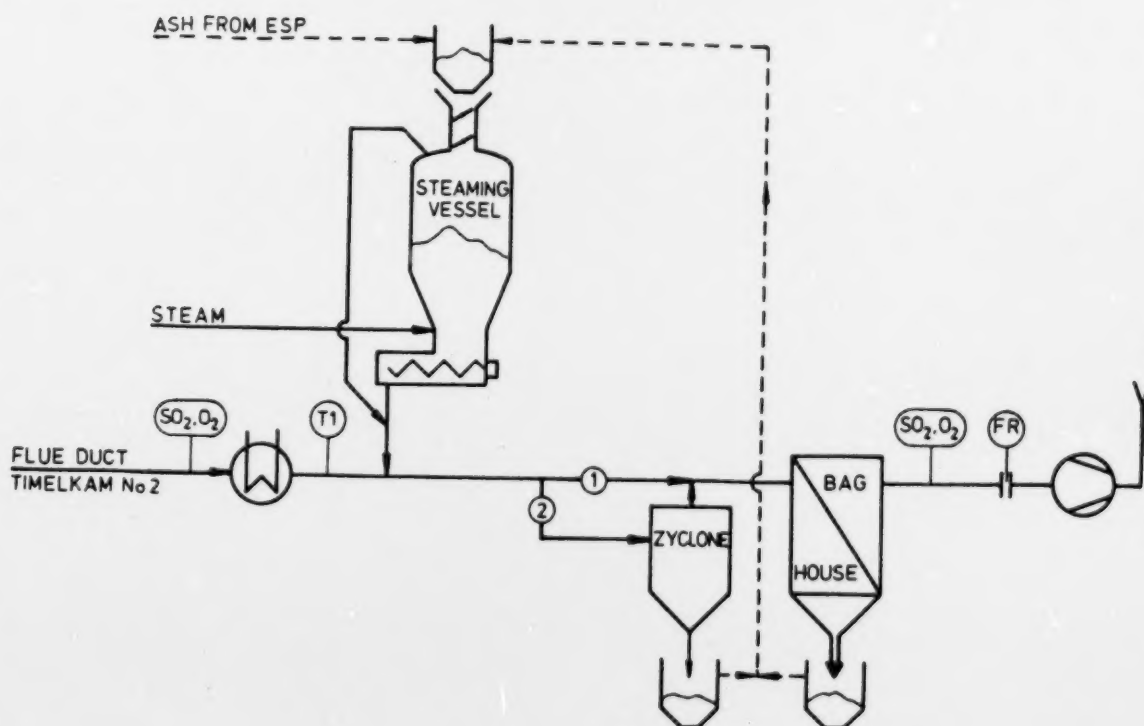


Figure 4. Pilot plant of Ash Recycle Activation Process.



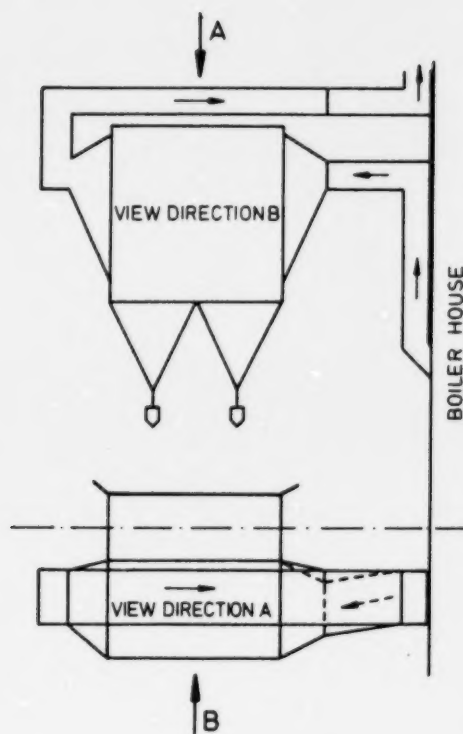


Figure 5. Present situation of ESP in Timelkam No 2.

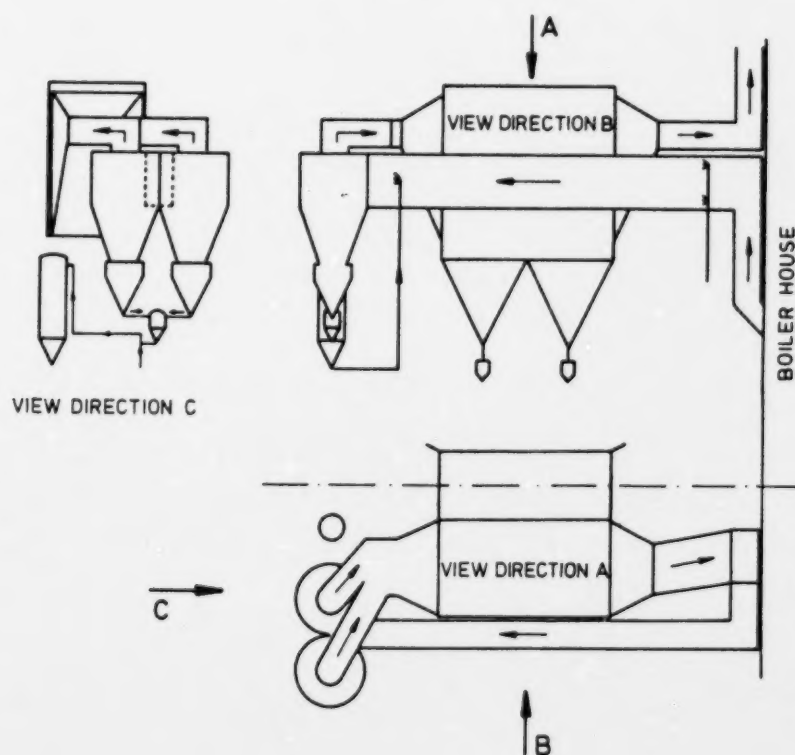


Figure 6. Concept for ARA and ESP in Timelkam No 2.

TABLE 1 INSTALLATIONS WITH FURNACE LIMESTONE INJECTION IN AUSTRIA AS OF AUGUST 1988

| PLANT NAME                          | OWNER                    | Plant erection year  | Instal-<br>l. year | Power<br>MWel    | LHV<br>kJ/kg              | Water<br>% | C o a l<br>Ash % | Sulfur<br>% | Burner<br>type               | Approx.<br>hours of<br>FLI<br>operation | Present<br>status                             |
|-------------------------------------|--------------------------|----------------------|--------------------|------------------|---------------------------|------------|------------------|-------------|------------------------------|-----------------------------------------|-----------------------------------------------|
| Voitsberg<br>No.2                   | ÖDK                      | 1956                 | 1982*              | 65               | 10500                     | 32to<br>38 | 15to<br>30       | 0.4 to 1.5  | 4 corner                     | 8000                                    | conserved,<br>if reactivated then<br>with FLI |
| St. Andrä<br>No.2                   | ÖDK                      | 1959                 | 1983*              | 110              | 9500-1300                 | 35         | 15to<br>25       | 0.5 to 1.2  | 4 corner                     | 11700                                   | FLI plus<br>sodiumbicarbonate                 |
| Voitsberg<br>No.3                   | ÖDK                      | 1983                 | 1983               | 330              | 10500                     | 32to<br>38 | 15to<br>30       | 0.4 to 1.5  | 6 corner                     | 8700                                    | wet FGD since 1986                            |
| Cogeneration<br>plant, Graz         | STENEAG<br>**            | 1964                 | 1984               | 3x18             | 12800                     | 36         | 12               | 0.4 to 1.2  | 4 wall                       | 55267                                   | 1 boiler on gas or<br>coal+FLI, 2 boilers FLI |
| Riedersbach<br>No.1                 | OKA<br>***               | 1969                 | Jan.<br>1985       | 55               | 10400-<br>12000           | 30         | 23.5             | 0.45to 1.9  | 4 corner                     | 10360                                   | FLI                                           |
| Timeilkam<br>No.2                   | OKA                      | 1962                 | 1985               | 60               | 9600<br>11300             | 38         | 15               | 0.4         | 4 corner                     | 14400                                   | FLI                                           |
| Riedersbach<br>No.2                 | OKA                      | 1986                 | 1986               | 160              | 10400<br>12000<br>(17000) | 30<br>(10) | 23.5             | 0.45to 1.9  | 4 corner                     | 2100                                    | wet FGD since 1986                            |
| Cogeneration<br>plant<br>Klagenfurt | Stadt<br>Klagen-<br>furt | 1948<br>1986<br>1958 | 1986               | 30+<br>50+<br>70 | 24700                     | 10         | 12               | 0.6 to 0.86 | 2 wall<br>4 wall<br>4 corner | 5000<br>5000<br>5000                    | meets future<br>limits with<br>FLI            |

\* Year of permanent installation  
 \*\* Steirische Wasserkraft und Elektrizitäts AG  
 \*\*\* Oberösterreichische Kraftwerke AG  
 + t/h steam

TABLE 2 COAL ANALYSIS OF OROSZLANY

|       |             |
|-------|-------------|
| Water | 14.24 %wt   |
| ash   | 46.33       |
| C     | 27.32       |
| H     | 2.15        |
| O     | 6.08        |
| N     | 0.24        |
| S     | 3.64        |
| LHV   | 8 792 kJ/kg |

TABLE 3 ASH ANALYSIS OF OROSZLANY

|                                              | without FLI     | with FLI      |
|----------------------------------------------|-----------------|---------------|
| loss on ignition                             | 1.14 - 0.96 %wt | 0.8 - 5.3 %wt |
| CO <sub>2</sub>                              | not measured    | 0.46 - 0.69   |
| combustible carbon                           | not measured    | 0.3 - 2.6     |
| SiO <sub>2</sub>                             | 45.64 - 49.0    | 38.2 - 41.9   |
| R <sub>2</sub> O <sub>3</sub>                | 24.5 - 24.95    | 22.1 - 24.5   |
| CaO                                          | 17.62 - 20.0    | 21.4 - 25.9   |
| MgO                                          | 1.03 - 1.8      | 1.1 - 2.09    |
| SO <sub>3</sub>                              | 2.45 - 2.5      | 4.4 - 5.4     |
| others (Na <sub>2</sub> O, K <sub>2</sub> O) | 2.23            | not measured  |

TABLE 4 SOME RESULTS FROM THE ARA PILOT PLANT AT TIMELKAM No 2

| RUN No | CABE<br>g/m <sup>3</sup> | SO <sub>2</sub> IN <sub>3</sub><br>mg/Nm <sup>3</sup><br>dry | CA/S | ESGR |
|--------|--------------------------|--------------------------------------------------------------|------|------|
| 18     | 24.9                     | 2 500                                                        | 15.9 | 47   |
| 21     | 37.9                     | 2 500                                                        | 24.3 | 52   |
| 29     | 16.4                     | 2 400                                                        | 10.9 | 37   |
| 31     | 33.4                     | 2 000                                                        | 26.0 | 55   |
| 33     | 33.2                     | 2 200                                                        | 24.1 | 50   |

4 YEARS OF OPERATING EXPERIENCE WITH DIRECT  
IN-FURNACE SO<sub>2</sub> REDUCTION AT THE 600 MWe UNIT  
OF PROVENCE POWER STATION

- Michel VANDYCKE, STEIN INDUSTRIE, FRANCE
- Bernard AURIC, CHARBONNAGES DE FRANCE, PROVENCE  
POWER STATION
- Jean-Louis MERRY, CHARBONNAGES DE FRANCE, CERCHAR

A B S T R A C T

A series of test conducted on a 50 MWe boiler led to the decision to adopt an economical direct in-furnace desulfurization system for the new 600 MWe unit of PROVENCE power station (south of FRANCE, near MARSEILLE).

This power station uses a quite unique sub-bituminous coal with a very high sulfur content (5.5 lb/MBtu) and such a high CaO content (50% of the ash) that some reduction of the potential SO<sub>2</sub> emission (30-40%) naturally occurs in the boiler.

Since October 1984, the desulfurization system by lime and limestone injection has been operating satisfactorily with a permanent total SO<sub>2</sub> capture higher than 60%.

The paper details the general characteristics of the desulfurization system, the conclusions of four years of operation and the improvements which have been carried out after the commissioning in order to increase the process efficiency.



## INTRODUCTION

PROVENCE power station is located in the south of FRANCE at a place called GARDANNE in PROVENCE.

This station uses a local coal mined by HOUILLERES DE PROVENCE, a company of the nationalized group "CHARBONNAGES DE FRANCE". The output of the mine amounts to about 1.5 million tons per year and a large fraction of it is used for electricity production.

This coal is a quite unique sub-bituminous coal, with a high ash content (20 to 30%) of which half or more is calcium oxide as this coal contains a significant amount of  $\text{CaCO}_3$ . Also it has a very high sulfur content, around 4% or more precisely 5.5 lb/MBtu - Figure 1.

The Station is now made of 2 units, one 250 MWe unit built in the seventies and one 600 MWe unit which was commissioned in October 1984.

The 600 MWe has been constructed by STEIN INDUSTRIE and it was initially authorized by the French administration without any desulfurization system but upon the condition that a very tall stack was to be installed (990 feet) to improve pollutant dispersion.

During the erection of this unit, the high sulfur level of GARDANNE coal gave rise to much concern regarding atmospheric pollution, despite the tall stack and some natural sulfur retention in the coal ash. After abandoning the idea of installing a Flue Gas Desulfurization system, which was considered excessively expensive, CHARBONNAGES DE FRANCE decided to investigate the Direct Desulfurization in the furnace with the assistance of its research center (CERCHAR) and of STEIN INDUSTRIE, in order to determine if it would be possible to reach the 60% sulfur retention now requested by the french administration.

A research and development program was then initiated in order to demonstrate the viability of this technique for the new 600 MW boiler that was already partially built.

## RESEARCH AND DEVELOPMENT PROGRAM

The first phase consisted of preliminary experimental furnace investigations just to check the main trends before large scale testing on a old 50 MWE boiler. This old unit was built by, STEIN INDUSTRIE in the fifties at the PROVENCE power station and is now out of service. It was tangentially fired and we had used it as a model for the design of the new 600 MW unit.

Firing GARDANNE coals of different ash content (therefore different Ca/S) permitted to monitor the natural retention effect. Various pulverized sorbents especially limestone  $\text{CaCO}_3$  and hydrated lime  $\text{Ca(OH)}_2$ , were injected into the furnace, either through the operating burners or above. These tests essentially demonstrated that the added limestone had the same desulfurization efficiency than the natural calcium carbonate contained in the GARDANNE coal, when hydrated lime injected above the flame had a much higher efficiency.

The conclusion was that the requested 60%  $\text{SO}_2$  capture could be reached with a reasonable lime/limestone injection and the success of this program led to the decision to adopt direct desulfurization for the new 600 MW unit.

The results of this research and development program have been presented at the first joint symposium on dry  $\text{SO}_2$  and simultaneous  $\text{SO}_2/\text{NO}_x$  control technologies EPA/EPRI - SAN DIEGO - NOV. 1984.

#### THE 600 MWE BOILER OF PROVENCE POWER STATION

The GARDANNE coal has a high fouling potential (risk of calcium sulfate deposits in the convection zone) due to its natural limestone content. Using STEIN INDUSTRIE experience with difficult coals, and particularly the experience derived from the operation of the old 50 MWe unit, we have designed the 600 MW boiler to minimize fouling, specially by increasing furnace size (by more than 60% in volume compared to other 600 MW units designed for imported coals), by installing more sootblowing capacity than usual (with rotary water lances in the furnace), by increasing the transverse spacing between the first heat exchanger tubes (which are platenized) and by selecting a higher excess air than usual (about 35%). Also, the tower type design avoids any risk of plugging the heat exchanger by loosened deposits (as, when they fall down, they go through larger and larger spacings between tubes) - Figure 2.

#### DESULFURIZATION PROCESS

The main design features of the desulfurization system are given below (see also the sketch Figure 3) :

- As half or more of the ash is calcium oxide, resulting of  $\text{CaCO}_3$  contained in the coal, Ca/S molar ratio varies from 1.5 to 2.5 for the coal alone.
- Limestone is added to the coal, upstream of the mills, in order to maintain, whatever the coal characteristics, a Ca/S ratio of 2.5 which was expected to result in about 45% sulfur capture.
- Pulverized hydrated lime is pneumatically injected into the furnace from the corners, above the upper burner elevation, in order to reach a total  $\text{SO}_2$  capture of 60% which was expected to correspond to a total Ca/S ratio of about 3. This lime injection is automatically controlled by unit load, plus fine tuning by stack  $\text{SO}_2$ .

Although limestone is much cheaper than hydrated lime, it was not considered to use limestone only, as it would have been necessary to use more of it, therefore increasing the solids in the flue gas above what was regarded as acceptable. Naturally the effect of the increased solid loading on the electrostatic precipitator as well as on the ash handling system, was taken into account (a fifth field was added).

The capital cost of this equipment (about 1.5% of the power station equipment cost) is naturally much lower than that of a conventional FGD system.

The various tests performed during commissioning led to the conclusion that the "natural" sulfur dioxide capture (due to limestone already present in the coal or added to the coal) is slightly lower than expected (a 45% capture corresponds to Ca/S about 3 instead of 2.5). The sulfur retention improves when load is decreased or when excess air increased.

The effect of lime injection is exactly as predicted. All these results are summarized on figure 4.

The desulfurization system is in commercial operation since October 1984, as the operating permit for this unit demands to maintain a total SO<sub>2</sub> capture higher than 60% and SO<sub>2</sub> content in flue gas always lower than 1540 ppm (dry gas, 11% CO<sub>2</sub>).

#### INVESTIGATION PROGRAM ON SO<sub>2</sub> REDUCTION OPTIMIZATION

An investigation program aiming at optimizing the system, especially at reducing the lime consumption, has been undertaken.

##### Lime injection tests at different locations

The idea was to research the optimum flue gas temperature at the lime injection level in order to improve the sulfation reaction.

The tests were performed with injection of lime from each corner and at 4 different levels, one after another (see figure 5a).

We observed that the lime efficiency decreases when the injection level increases. For example, in the same operating conditions, the SO<sub>2</sub> removal increment (due to the lime) over the "natural" absorption efficiency was :

- 11% with injection at 199 and 215 feet level
- 6% with injection at 235 feet level
- 2% with injection at 251 feet level

This could be caused by reduction of the particle residence time at the temperatures which are conducive to the sulfation reaction when the injection level increases. In addition, the mixing between gas and lime particles is probably very poor for the upper levels.

## Optimization of lime and gas mixing

### Full scale testing

The velocity of the lime transport air at injectors was increased from 92 feet/sec to 213 feet/sec, by a reduction of the injector section.

We observed no significant improvement of lime efficiency and we decided to study the mixing on a reduced scale 3 D model.

### 3 D model - Visualization of the sorbent jets

The tests were conducted on a water model which simulates the geometry of the tangentially fired 600 MW unit (scale 1/40, figure 6).

The model was built of clear plastic and represented the entire furnace from the hopper to the first heat exchanger (HTS). The lime injection was visualized with coloured liquid and a grid of sample probes was installed on a cross section at the equivalent of 28 feet above injection level (199 feet).

The coloured liquid flow rate was maintained constant and we tested the following configurations :

- injector near each corner; 2 carrier air velocities and 3 sorbent jets orientations (see figure 5b).
- injector at the center of each wall (perpendicular) and 2 carrier air velocities.

In a first screening test we conducted flow visualization at simulated full load furnace operating conditions. In a second screening we selected the configurations leading to the best mixing and performed measurements of coloured liquid concentrations and velocity profiles.

The best mixing was obtained with the injectors at the center of each wall and with the low transport air velocity. When the velocity is increased, the concentration in the center of the furnace becomes too high.

With the injectors in the vicinity of the corners, the mixing is always very poor, specially in the center of the furnace (figure 7).

These investigations led to the decision to adopt the new injection location for the 600 MW units.

### Sorbent characteristics evaluation in a laboratory plug flow furnace

Sorbent characteristics evaluation tests were carried out in a specially build plug flow furnace by CERCHAR at VERNEUIL Laboratory. Lime and limestone samples from PROVENCE power station were then ranked by desulfurization efficiency.



The furnace is composed by four parts :

- a synthetic flue-gas preparation assembly,
- a preheater used to heat gases up to 2190°F,
- a 6.5 feet length and 2.4 inches internal diameter reactor tube where the additive is injected into the gas flow. The reactor is heated up to 2190°F
- a sampling apparatus with an insulated semi-cooled probe and a filtration case.

In the reactor tube, the gas velocity is such that the flow is laminar and the gas residence time is about of one second or less.

By varying the distance between the additive injection point and the probe, we are able to increment the residence time by 50 ms each time. The evolutions of additive specific area and desulfurization rate, in terms of residence time, were measured from 50 ms to 700 ms. We plotted these values, showing that the maximum specific area is situated near 150 ms. The desulfurization speed (i.e. between 350 ms and 700 ms) is directly in relation with the chemical impurities of sorbent (Fe, Mg....).

During a second period, we limited the parameters exploration and set fixed values for residence time (700 ms), temperature (2190°F) and SO<sub>2</sub> concentration (2300 ppm), just varying the Ca/S ratio. Some results with lime samples are shown on figure 8. The same procedure applies to limestone as well. According to the representativity of samples, it is roughly possible to evaluate sorbent quality, and chose sorbent suppliers.

#### Other investigations

In order to reduce the operating cost some tests were performed by using pulverized limestone instead of hydrated lime for furnace injection. The improvement of SO<sub>2</sub> removal efficiency due to the new location of the lime injectors, could give us the possibility of using limestone, in spite of its lower efficiency. The first results are promising but more investigations are necessary.

#### CONCLUSION

Since October 1984, the desulfurization system by lime and limestone injection has been operating satisfactorily with a permanent total SO<sub>2</sub> capture higher than 60%. The improvements which have been made during the last 4 years, give a better knowledge of the system. Naturally, such a process can be used elsewhere, when the boiler design is suitable, if partial desulfurization is acceptable.

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|                                                                   | Base coal | High ash coal |
|-------------------------------------------------------------------|-----------|---------------|
| PROXIMATE, WT % (including CO <sub>2</sub> )                      |           |               |
| MOISTURE                                                          | 12.0      | 10.7          |
| VOLATILE MATTER                                                   | 38.2      | 38.4          |
| FIXED CARBON                                                      | 27.0      | 22.9          |
| ASH                                                               | 22.8      | 28.0          |
| ULTIMATE, WT % (including CO <sub>2</sub> )                       |           |               |
| MOISTURE                                                          | 12.0      | 10.7          |
| ASH                                                               | 22.8      | 28.0          |
| CARBON                                                            | 45.7      | 41.4          |
| HYDROGEN                                                          | 3.1       | 2.9           |
| TOTAL SULFUR                                                      | 4.1       | 3.9           |
| NITROGEN                                                          | 0.9       | 1.2           |
| OXYGEN (Diff)                                                     | 11.4      | 11.9          |
| HIGHER HEATING VALUE, KCAL/KG<br>BTU/LB                           | 4360      | 3790          |
|                                                                   | 7850      | 6820          |
| ASH, WT %                                                         |           |               |
| CaO                                                               | 51.0      | 59.9          |
| Ca/S MOLAL RATIO                                                  | 1.6       | 2.5           |
| SULFUR CONTENT, KG/10 <sup>6</sup> KCAL<br>LB/10 <sup>6</sup> BTU | 9.4       | 10.3          |
|                                                                   | 5.2       | 5.7           |

FIGURE 1 : TYPICAL ANALYSIS OF GARDANNE COAL (AS FIRED BASIS)

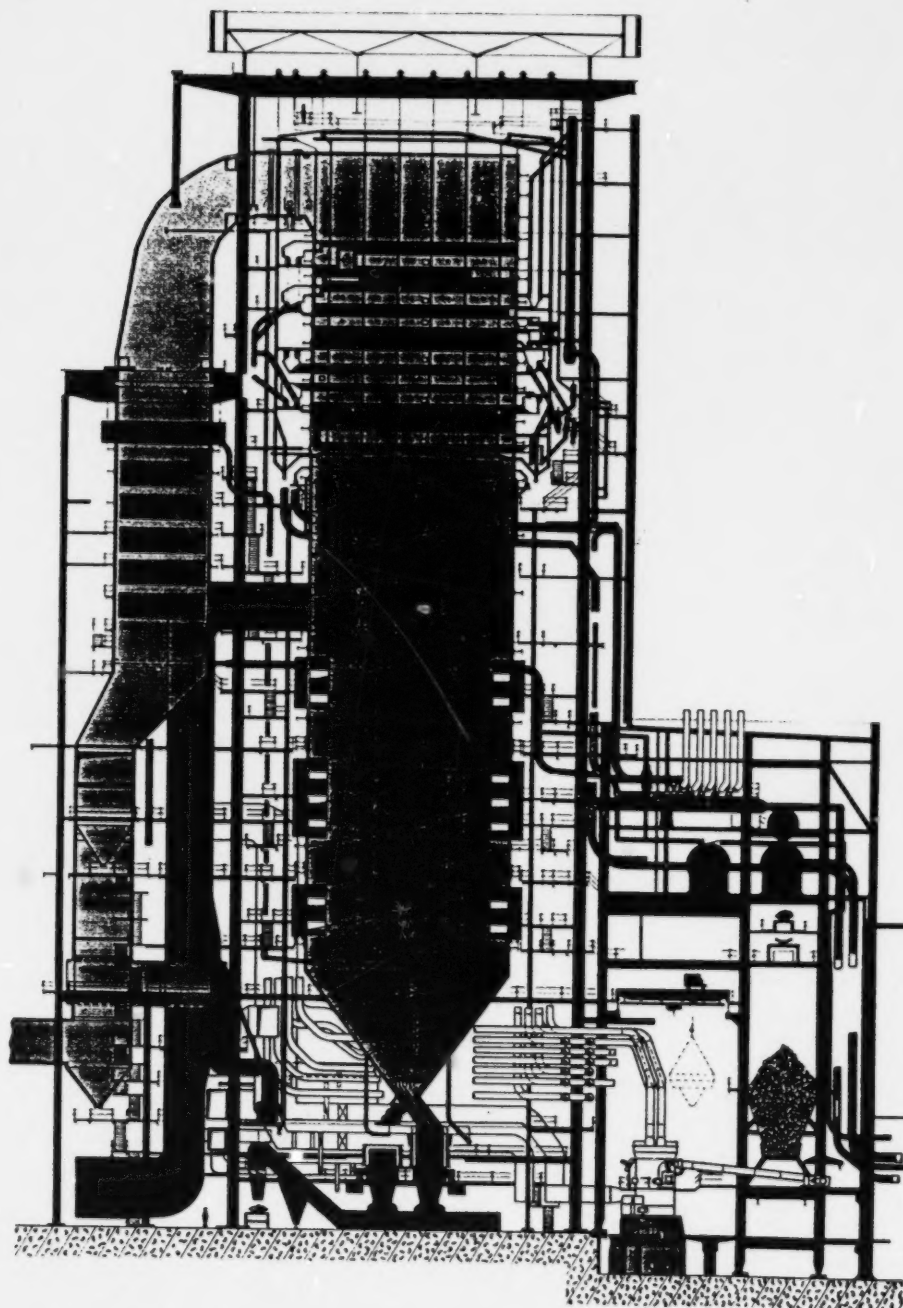


FIGURE 2 : CROSS SECTION OF THE 600 MW MONOTUBE STEAM GENERATOR  
PROVENCE UNIT NO 5.

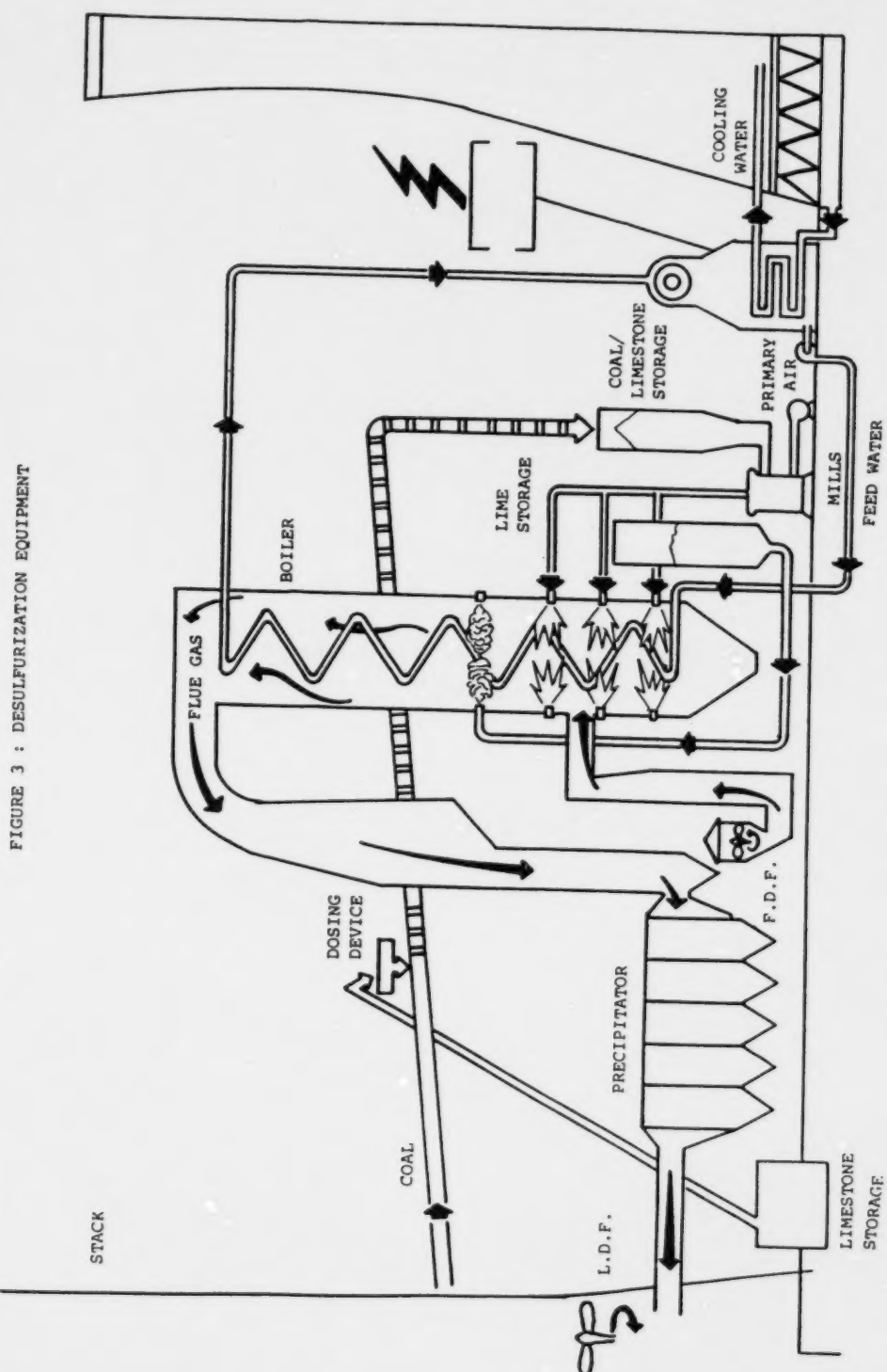


FIGURE 3 : DESULFURIZATION EQUIPMENT



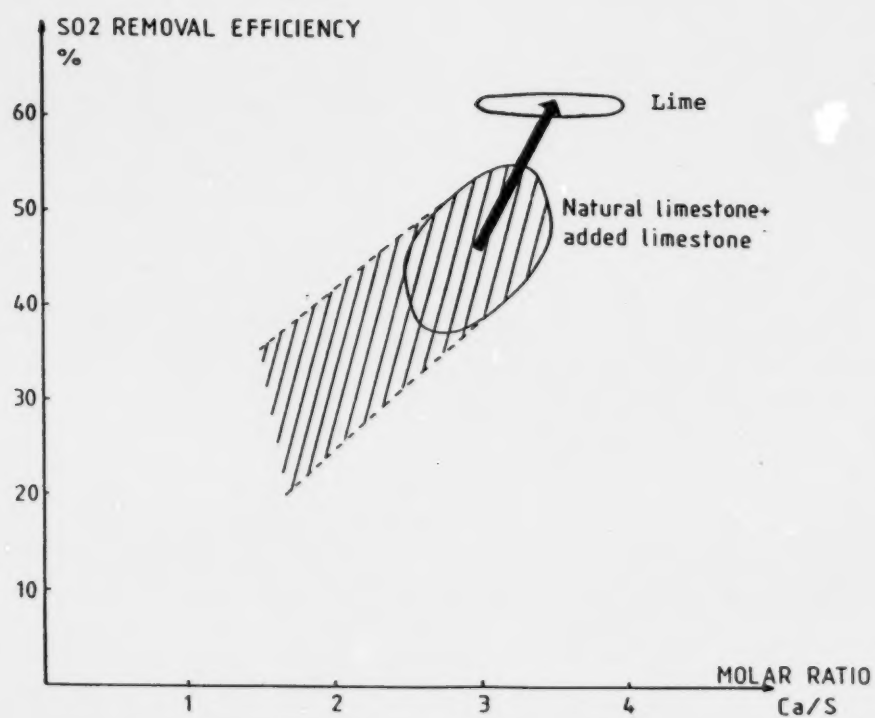


FIGURE 4 : DESULFURIZATION RESULTS

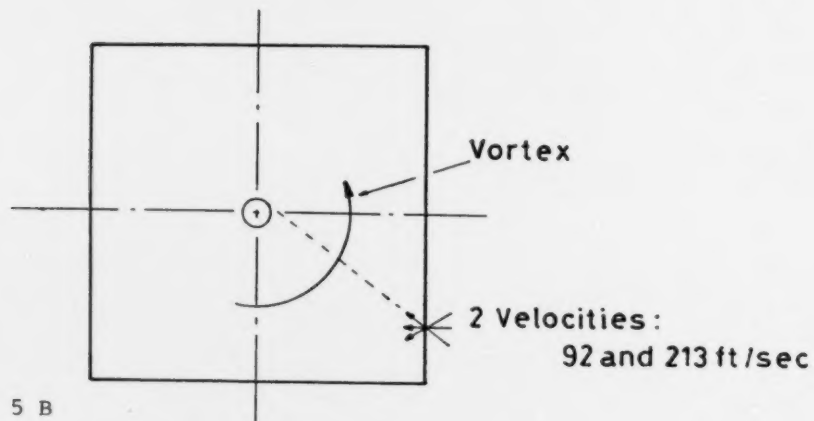
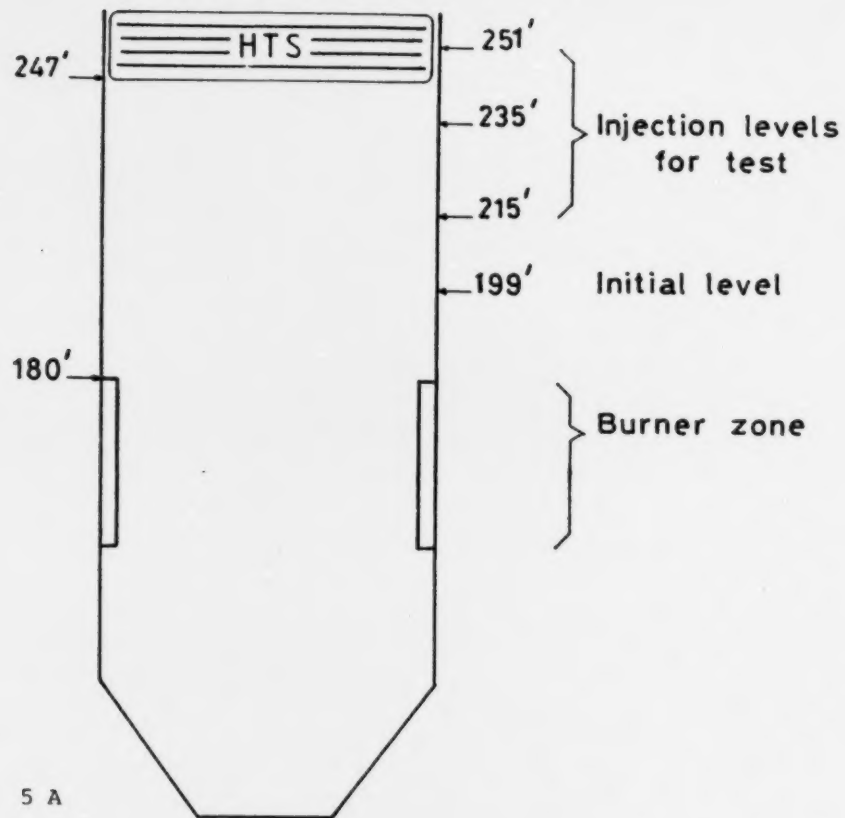


FIGURE 5 : LOCATION OF ADDITIVE INJECTION

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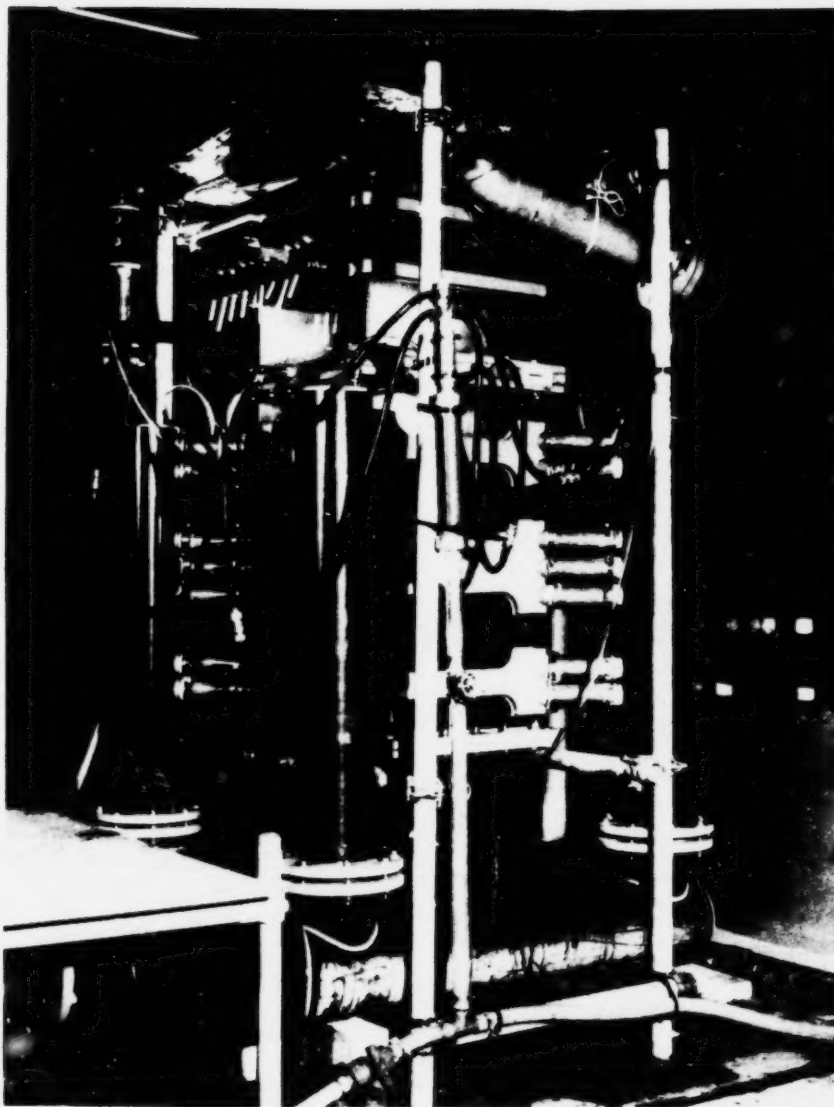
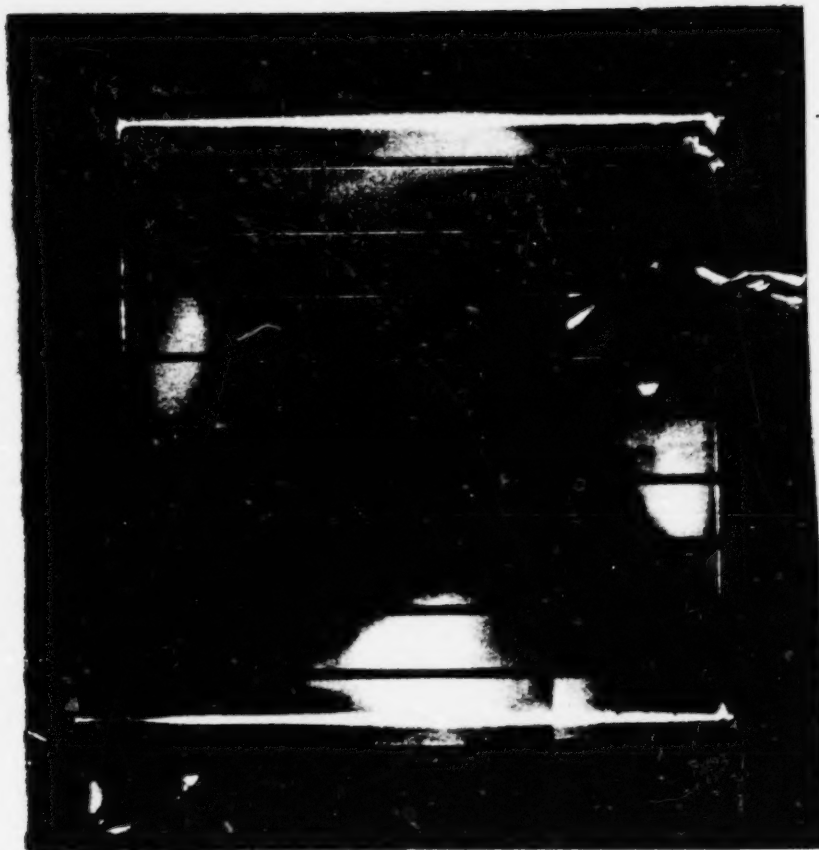


FIGURE 6 : 3-D SIMULATION MODEL



POOR MIXING, LIME INJECTORS IN THE VICINITY OF  
THE CORNERS

FIGURE 7 : 3-D MODEL : VISUALIZATION OF SORBENT JETS



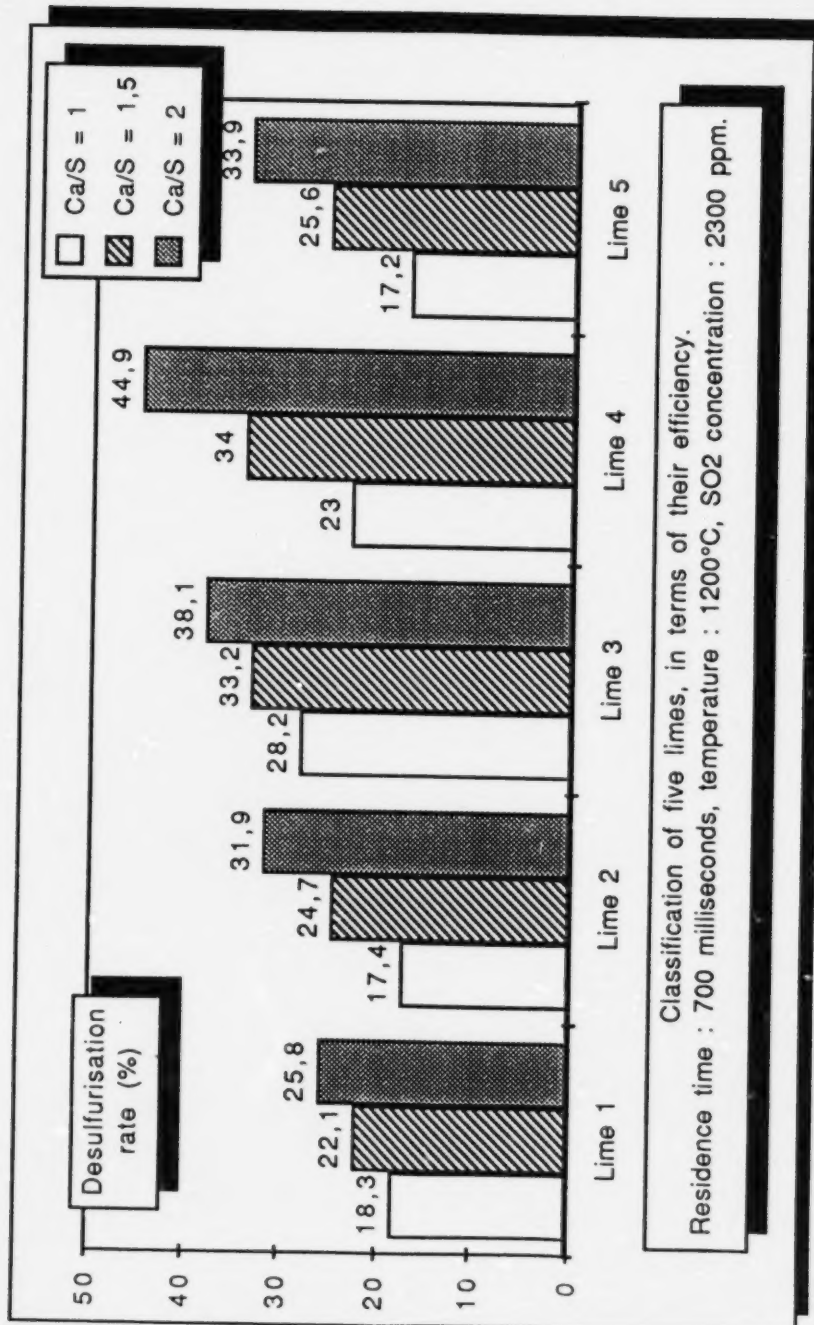


FIGURE 8

## SESSION 4B: INTEGRATION/BYPRODUCT UTILIZATION

Chairman, Michael W. McElroy, EPRI

### IMPROVING SLUDGE DEWATERING IN MAGNESIUM-ENHANCED LIME FGD SYSTEMS

by

Lewis B. Benson  
Dravo Lime Company  
Pittsburgh, PA 15222

Dr. Alan Randolph  
University of Arizona  
Tucson, Arizona 85721

Jim Wilhelm  
Codan Associates  
Sandy, Utah 84092

#### ABSTRACT

EPRI encourages electric utilities with wet FGD systems to improve their operation by first understanding the chemical processes that occur in wet FGD and then applying this understanding to their systems.

This paper describes application of this approach to improve sludge dewatering in magnesium-enhanced lime wet FGD systems. Earlier work has shown that a key to improving performance of thickeners and filters in these systems is to adjust scrubber operating conditions to produce larger sludge particles.

First described are the purpose and results of laboratory crystallization tests conducted by the University of Arizona for Dravo Lime Company which confirmed scrubber operating conditions that increase particle size of calcium sulfite crystallized in magnesium-lime systems.

Next described are results of successful application of the findings of the laboratory tests to increase sludge particle size and improve sludge dewatering in a 300 MW wet FGD system using magnesium-enhanced lime. Thickener underflow solids concentration increased from 25-30 wt.% to 35-40 wt.% and centrifuge cake solids concentration increased from 45-50 wt.% to 56-65 wt.%. Discussion includes operating cost reductions caused by these improvements.

Finally the implications of these findings for sizing of dewatering equipment for future magnesium-enhanced lime FGD systems are discussed.

## INTRODUCTION

Wet flue gas desulfurization using magnesium-enhanced lime as reagent is widely practiced in the eastern U.S. Such lime-based systems achieve high sulfur dioxide ( $\text{SO}_2$ ) removal efficiencies and high reliability (1). These systems convert  $\text{SO}_2$  absorbed from flue gas to calcium sulfite hemihydrate.

Calcium sulfite sludge produced by magnesium-enhanced lime systems has been more difficult to settle and dewater than sludge produced from limestone-based systems. It has been suggested that magnesium inhibits calcium sulfite crystal growth, but little work had been performed to determine what factors actually influence crystal size in magnesium-lime FGD systems or to find scrubber operating conditions which improve crystal size in these systems.

The key to improving dewatering in magnesium-enhanced lime FGD systems is to find scrubber operating conditions which cause larger sludge particles to be produced. The performance of FGD dewatering equipment is determined largely by the size of the sludge particles exiting the scrubbers. Production of larger particles will cause an increase in the concentration of solids in thickener underflow and in dewatered cake in an existing scrubber and will allow smaller thickening and dewatering equipment to be selected for a new system.

Dravo Lime began study of means to increase crystal size and improve dewatering in these systems in 1985. An earlier paper (2) reported results of limited full-scale tests performed at the suggestion of Codan Associates to improve dewatering in magnesium-lime systems. The data showed a trend toward production of larger, better settling sludge particles when the scrubber recycle tank was operated with a slurry solids concentration lower than design.

This finding was surprising because it was contrary to conventional wisdom in the FGD industry. Limestone, lime and magnesium-enhanced lime FGD scrubbers are designed to operate with 10-15 wt.% slurry solids concentrations. Limestone systems are designed this way because a high limestone concentration in the slurry is required when it contacts flue gas to achieve desired  $\text{SO}_2$  removal. High design scrubber slurry solids concentration apparently was carried over to magnesium-enhanced lime-based systems.

The high slurry solids concentration used in design of wet lime and limestone systems results in long residence times for sludge particles in the scrubber. High slurry solids concentration is achieved by discharging slurry from the scrubber recycle tank to the dewatering system at a low rate; the rate is such that sludge particles spend an average of 12 to 24 hours in the tank. It has been assumed that this is beneficial for dewatering because one might expect longer residence time to give sludge particles more time to grow and so to grow larger. (Moderately longer residence times have proved beneficial in some limestone-based systems and in some systems that force oxidize captured  $\text{SO}_2$  to gypsum.)

Excessively long particle residence times are likely destructive to particles produced in magnesium-lime systems. The crystals that form are rosette-shaped and plates that comprise the crystals are relatively thin. This fragile structure makes the crystals susceptible to breakage by recycle tank agitators and by recirculation through scrubber recycle pumps. Operation at design scrubber suspended solids concentration and liquid-to-gas-ratio cause particles to recirculate hundreds of times. High recirculation also causes more mechanical energy to be delivered to each element of slurry, increasing potential for nucleation.

Reducing particle residence time to reduce particle breakage would also likely change the conditions that control growth of crystals in the recycle tank. Literature on FGD crystallization suggested that under certain conditions one could expect shorter residence time to produce larger particles (3,4), as had been observed in the above-noted full-scale tests. Average crystal size produced in a well-mixed FGD recycle tank is set by the nucleation rate (the rate at which new, very small particles precipitate) and the particle growth rate (the rate at which particles already present grow). If a decrease in residence time causes the nucleation rate to decrease, an increase in average sludge particle size should result. Similarly, even if a decrease in residence time causes both the nucleation rate and growth rate to increase, an increase in average particle size will result if the growth rate increases more rapidly than the nucleation rate.

## LABORATORY TESTS TO STUDY CRYSTAL GROWTH AND NUCLEATION

### PURPOSE OF TESTS

Although the full-scale data collected earlier showed a trend toward larger particle size with lower scrubber slurry concentration, Dravo Lime felt the need to conduct a controlled laboratory study that could verify this trend and to study how changes in residence time effect crystal nucleation and growth before conducting further full-scale tests. Such lab testing would, for example, allow scrubber liquor magnesium concentration to be held constant while particle residence time was changed. This compares with conducting full-scale tests where sometimes liquor magnesium concentration falls with suspended solids concentration. In this case, it is difficult to attribute an increase in particle size only to decreased suspended solids concentration and residence time.

In addition, these lab tests would allow better understanding of the independent effects on particle size of several other scrubber operating conditions: pH, agitation by the crystallizer agitator, agitation by recycle pumps, and scrubber loading. One goal of the tests would be to determine whether each of these conditions separately caused an increase or a decrease in particle size.

Dravo Lime commissioned Dr. Alan Randolph of the University of Arizona to conduct laboratory crystallization tests that would model conditions in the recycle tank of a scrubber using magnesium-enhanced lime. (A well-mixed tank in which crystallization occurs and from which representative slurry is withdrawn is called a mixed-suspension mixed-product removal, or MSMR, crystallizer.) The tests were conducted using a crystallizer and other equipment used in earlier laboratory FGD crystallization tests conducted for EPRI (5). A scrubber liquor magnesium concentration of about 5000 mg/liter was used in all the tests. Use of the same test equipment and similar conditions as in earlier work would allow growth and nucleation rates to be determined for the magnesium-enhanced lime system that could be compared with rates obtained earlier.

## OUTLINE OF TEST PROGRAM

A series of MSMR runs in an 11-liter draft-tube-baffled crystallizer was planned to test residence time, agitation rate, slurry density (as varied by feed to circulating liquor rate), pH, rate of make, absorption of SO<sub>2</sub> in the precipitating tank vs. an external absorber and the flow rate of slurry to the external absorption tank. Figure 1 shows the crystallizer apparatus. All of these changes were compared to a base run with conditions shown in Table 1.

The above variables were changed one by one from the base. The following is a summary of responses. The overall responses were confirmed by correlating calculated growth and nucleation rates with a power-law kinetics model proposed by Keough (6), namely:

$$B^0 = K_N G^{0.63} M_T^{2.6} \epsilon$$

where:  $B^0$  = nucleation rate, nuclei formed per liter per minute

$K_N$  = nucleation rate constant

$G$  = particle growth rate, microns/minute

$M_T$  = slurry solids concentration, grams/liter

$\epsilon$  = power transferred to the slurry by the agitator per weight of slurry, watts/kg

The data from the present study fit the above power-law model (Figure 2), but nucleation was uniformly lower (i.e. larger particles were produced). This may have been because Keough used a concentrated solution of NaHSO<sub>3</sub> as source of SO<sub>2</sub> feed rather than direct absorption of SO<sub>2</sub> as in this study or because of the enhanced Mg content of the present study.

The following is a summary of the responses obtained by perturbing the given variables from the base case:

1. **Particle residence time:** Shorter residence times produce a larger particle size as predicted from Keough's data.
2. **Rate of make:** No effect except size degeneration if higher slurry density resulted.
3. **Increased agitation:** Produced smaller particles. Increased nucleation proportional to  $\epsilon$ , power per unit mass of suspension.
4. **pH reduced from 7 to 6:** Degeneration of particle size.
5. **Recirculation of slurry thorough centrifugal pumps:** Large degeneration of particle size
6. **Internal vs. external SO<sub>2</sub> absorption:** No effect. (Some early tests were conducted to compare the effect on particle size of SO<sub>2</sub> absorption in an external absorber as shown in Figure 1 compared to absorption in the crystallizer. No difference in particle size was observed, so the balance of tests were conducted with absorption in the crystallizer.)



## DESCRIPTION OF TEST PROCEDURES AND CONDITIONS

The Thiosorbic® scrubbing process was carried out in an 11.4 liter stainless steel crystallizer at 50°C. A solution whose chemical composition equaled that of thickener return liquor from an actual magnesium-lime FGD system was fed to the crystallizer; it contained 5000 mg/liter magnesium. The concentration of other constituents is given in Table 2. A 5 lb. gas cylinder was used as the sulfur dioxide (SO<sub>2</sub>) gas source. Slaked magnesium-enhanced lime (Thiosorbic® Lime) containing 6 wt% MgO was added to react with the SO<sub>2</sub> and precipitate calcium sulfite.

The feed solution, representing thickener return liquor in Dravo's actual scrubbing process, was made up the evening prior to the experiment. The reagents used to make the feed solution were MgSO<sub>4</sub>·H<sub>2</sub>O, MgCl<sub>2</sub>·7H<sub>2</sub>O, Mg(OH)<sub>2</sub> and 6% sulfurous acid solution. The concentrations of the magnesium compounds and conditions used are given in Table 2. The feed solution was kept at 50°C throughout the experiments.

The crystallizer was stirred using an 8.5 cm marine propeller. The crystallizer was 24.5 cm in diameter and contained a 9.0 cm (diameter) draft tube with four baffles (all stainless steel). The crystallizer was heated using glycol pumped from a constant temperature bath to copper coils surrounding the outside of the crystallizer. SO<sub>2</sub> gas was bubbled into the crystallizer solution by two methods. The first was to bubble the gas directly into the crystallizer and the second was to bubble the SO<sub>2</sub> into an absorption tank. Both of these methods are shown in Figure 2. The bubbler used was a coarse sintered glass filter. Nitrogen was fed with the SO<sub>2</sub> to dilute the SO<sub>2</sub> around the bubbler exit, preventing fouling of the glass frit.

In these tests SO<sub>2</sub> is absorbed directly into a calcium sulfite slurry to precipitate CaSO<sub>3</sub>·1/2H<sub>2</sub>O crystals. In this well-mixed process, Dravo's lime [Ca(OH)<sub>2</sub> with some Mg(OH)<sub>2</sub>] is introduced into the DTB stirred vessel at a fixed rate. SO<sub>2</sub> diluted to 8 mol.% with N<sub>2</sub> is absorbed into the mixture to maintain the pH at 7.0. CaSO<sub>3</sub>·1/2H<sub>2</sub>O crystals precipitate and are withdrawn as slurry discharge. Ten runs in this system were made to study precipitator residence time, pH, rate-of-make and rpm.

Two additional precipitator runs were made. One was to introduce a high rpm centrifugal circulating pump between the crystallizer and an external SO<sub>2</sub> absorber to study the effect of scrubber slurry recirculation on particle size. The second test was to study the effect of a higher liquor sulfite concentration (accomplished by adding more sulfite and slightly less sulfate to the simulated thickener return liquor) than in earlier runs.

## TEST RESULTS

The addition of a high-volume circulation of slurry between precipitator and absorber (using two 3,400 rpm Eastern D-11 circulating pumps in series) degenerated particle size from a previous average of 32 microns in the base case to less than 22 microns. In addition, a fines distribution of approximately 11 microns average was found in the presence of the 22 micron crystals. (The sizes reported above are the weight average particle size, calculated as four times the negative reciprocal slope of the population density-size plot per conventional MSMPR crystallizer theory. A Particle Data Inc. zone particle analyzer was used to determine particle size.)

Figure 3 shows two distinct straight line semi-log population plots as the distribution from Run 11. As these two distributions obviously overlap, the true mass mean of the entire crystal distribution would be somewhere between 11 and 22 microns.

The result of adding the slurry recycle was devastating to particle settling rate, which increased from a few minutes to nearly an hour. This result matches the size degeneration and slow settling rate observed for sulfite particles in commercial lime FGD units with large absorber/precipitator recirculation rates. It is believed that the two crystal distributions observed are composed of (a) normal-growing crystals which are somewhat smaller than average in size because of the higher secondary nucleation rate caused by slurry circulation, and (b) a fines distribution of intrinsically slow-growing crystals created from nuclei attrited from the larger distribution and/or by conventional secondary mechanisms. The phenomenon of nuclei created in the same system with the same supersaturation, but growing at different rates, is referred to in the literature as growth rate dispersion and has been widely reported, e.g. Berglund and Larson (7), and Randolph (8.)

In Run 12, higher sulfite levels gave 33 micron crystals, slightly larger than the average at lower sulfite concentrations. A high supersaturation of  $\sigma = 2.96$  was calculated for this run using a species distribution model provided by Dravo.

The crystal growth rates and supersaturations for Runs 11 and 12 reasonably followed the growth rate-supersaturation correlation,  $G = k_1\sigma + k_2$  obtained from earlier runs. Measured and predicted growth rates for Runs 11 and 12 are shown in Table 3. Growth data from Runs 11 and 12 essentially fit the correlation proposed from the first 10 runs. Run 11 would have been in closer agreement if crystals from the slow-growing segment of the population plot had been included in calculation of the growth rate.

Overall, the data supports the following conclusions which have important implications for operation and design of full-scale magnesium-enhanced lime FGD systems:

1. Produced larger particles with shorter residence time in support of earlier observations from full-scale tests.
2. Shear by centrifugal recycle pump produced a large decrease in size and fines with a lower growth rate.
3. The nucleation and growth rates derived from the data support the earlier observation of larger particles at lower residence times, higher supersaturations, and suggest high sensitivity of nucleation rate to slurry solids concentration.

## FULL-SCALE TESTS USING RESULTS OF LAB TEST PROGRAM

### INTRODUCTION AND PURPOSE

The results of the lab tests supported the earlier finding that particle size could be improved by reducing residence time and minimizing the number of times particles pass through recycle pumps. Importantly, these factors affected particle size independent of liquor chemistry.

Dravo Lime discussed these results with personnel from a utility using magnesium-enhanced lime in their FGD system and suggested testing to increase particle size and determine the resulting improvement in sludge dewatering.

The station at which testing was conducted is 300 MW and burns on average a 2.75% sulfur coal. Sludge from two operating spray-tower scrubber modules flows to two 100 foot diameter thickeners to concentrate the sludge. Thickened sludge is dewatered in a Sharples decanting centrifuge (model PM 75000) before it is mixed with flyash and lime and landfilled.

Sludge particle size exiting the scrubbers prior to testing was usually 15-19 microns weight average size (determined by a Leeds and Northrup Microtrac particle size analyzer). Thickener underflow solids concentration averaged 25-30 wt.%. Centrifuge cake solids concentration averaged 45-50 wt.%.

The goal of testing was to improve crystal size and subsequently thickener underflow concentration and centrifuge cake solids content by reducing particle residence time in the scrubber recycle tanks and reducing the number of times particles pass through recycle pumps.

### TEST CONDITIONS

A comparison of scrubber operating conditions before testing and during testing shows how this was accomplished. Before tests were conducted, a scrubber was normally operated at full load with 3 or 4 recycle pumps on, equal to a liquid-to-gas-ratio of 45 or 60, respectively. (Liquid-to-gas-ratio or L/G is defined as U.S. gpm of slurry per 1000 acfm of flue gas at scrubber outlet conditions, saturated with water at 125 F.) The scrubber recycle tank was kept 80% full of slurry with a volume of 192,000 gallons. Suspended solids concentration in the slurry was controlled at 6 wt.% by controlling the rate at which of thickener overflow was added to the scrubber and blowing down the slurry to the thickener to maintain level in the tank. At full load the blowdown rate was about 450 gpm.

These conditions, prior to testing, resulted in an average particle residence time of 7.1 hours in the scrubber before blowdown to the thickener. Particles circulated through recycle pumps an average of 89 times before blowdown to the thickener when 4 recycle pumps were on; average residence time of slurry in the recycle tank before recycle to the scrubber sprays was 6.4 minutes.

Scrubber operating conditions were changed to test reduced particle residence time and recirculation as follows: Each scrubber was operated with 2 recycle pumps on, equal to a L/G of 30. The level of slurry in the recycle tank was reduced to 139,200 gallons or 58% full. Blowdown of scrubber slurry was increased to 920 gpm and held at this rate, which reduced suspended solids concentration to a maximum of about 3 wt.%.

These conditions reduced average particle residence time to 2.5 hours before blowdown to the thickener. The average number of times a particle circulated through a recycle pumps was reduced to 22. Recycle residence time was rose slightly to 6.9 minutes. (Recycle residence time changed little because both the recirculation rate and the volume of slurry in the recycle tank were reduced.

Some short-term testing began in November 1987. During November through mid-February 1988, several week-long tests were conducted to study the effects of individual conditions like the number of recycle pumps

operating, slurry level, and pH. These conditions were compared one at a time by maintaining different conditions in each of the two operating modules. Data was collected on scrubber solids particle size, scrubber suspended solids concentration, scrubber liquor chemical composition, and thickener underflow solids concentration. These short-term tests confirmed that there was no detrimental effect on scrubber operation caused by reducing residence time to 2.5 hours or reducing the number of operating recycle pumps to 2.

## TEST RESULTS

Average particle size at low particle residence times increased to 20-23 microns. Figure 4 shows the observed effect of residence time on particle size during one of the tests. A drop in scrubber pH to 6 caused a drop in particle size as in the laboratory tests; a later test to duplicate this did not show this effect as clearly, perhaps because plant load decreased during this test.

Thickener underflow solids concentration increased significantly even during the period when weekly tests were being conducted. Figure 5 shows a plot of thickener solids during November and December. Solids concentration increased from 24-30 wt.% early in the test period to 30-43 wt.%, with most of the measurements showing solids concentrations above 35 wt.%. During station operation at full MW load, one thickener contained underflow concentrated to 38-42 wt% nearly every day.

Because of the improvement in dewatering that occurred during testing, both scrubbers continue to be operated at low particle residence times in normal practice. Figure 6 shows a plot of monthly average thickener underflow concentration in one of the thickeners and maximum concentration for each month for a typical period in 1986 and for the period following the beginning of testing. Thickener underflow concentration averaged above 37 wt% for three of the four months following the beginning of testing, with concentrations as high as 43 wt.% compared to 25-30 wt% in the earlier period. Centrifuge cake solids concentration improved also as shown in Figure 7. Centrifuge cake solids improved from 48-50 wt% in an earlier pre-test period to 53-56 wt% during short-term testing to 56-58 wt% with regular operation of the scrubbers in the low-residence time mode with measurements as high as 65 wt%.

## IMPROVEMENTS IN OPERATION RESULTING FROM IMPROVED DEWATERING

Improvement in particle size and sludge dewatering greatly reduced the need for polymers to aid dewatering. Prior to testing, polymer was added to the centrifuge feed to about 75-100 ppm to reduce carry-over of fine particles in the centrate. Polymer addition is no longer required. Higher thickener underflow concentrations reduced the volume of slurry to be centrifuged each day. Daily sludge centrifuging time fell from 13 to 9 hours, resulting in a reduction in clean-up and maintenance in the centrifuging and sludge treatment area.

## IMPACT OF INCREASED SLUDGE PARTICLE SIZE ON THICKENER AND FILTER SIZING FOR NEW SYSTEMS

### STRONG DEPENDENCE OF SIZING ON PARTICLE SIZE

Dewatering rates and concentrations of solids achieved in various dewatering devices are functions of particle size distribution of the sludge. While there are some theoretical equations that predict the relationships, they are only suitable for application to ideal systems with regular, uniform particle shapes that are incompressible. Most filter and thickener sizing is based either on previous experience with similar sludges or on actual bench-scale measurements of thickening and filtration characteristics.

Full-scale and bench-scale sizing criteria have been developed for wet lime scrubbing systems. As expected the data indicate that smaller thickeners and filters are required for systems that produce a larger particle size distribution. Thickener underflow concentrations and concentrations of solids in the filter cake are also improved as the particle size distribution increases.

Several methods have been developed for correlating dewatering characteristics with particle size distribution. One such method is to use a modified fineness modulus to represent the particle size distribution. In this paper the fineness modulus is equal to the sum of the cumulative percentages finer than 25 microns, 15 microns, 10 microns, and 5 microns, divided by 100. The modulus adds the percentage of the finest particles up to 4 times, while it excludes the particles larger than 25 microns, and counts the mid-size particles only once or twice. The larger the fineness modulus, the finer the particle size distribution. This modulus has been found to correlate the dewatering characteristics as shown in Figures 8-11.

Figure 8 is a plot of filter cake solids concentration as a function of fineness modulus. When the scrubbers are operated to produce smaller particles (as evidenced by a large fineness modulus), the filter cake solids concentrations drops as shown in the figure.



It should be noted that filter cake solids concentration can vary by changing cake drying time, cake thickness, vacuum levels, and vacuum air flow rates. The values plotted in the figure are typical design values based on bench-scale test data.

Figure 9 is a plot of filtration rate as a function of fineness modulus. Again, the filtration rate is a function of other variables in addition to particle size distribution. The values plotted are for a typical cake thickness of 3/8 to 1/2-inch on a drum filter with approximately 33% submergence with a feed solids concentration produced by a typical thickener as described below.

The data indicate that there is a predictable relationship between particle size distribution and filtration characteristics. When the scrubbers are designed and operated to produce the largest particle size distribution, the filtration rate can be over 200 lb/hr/sq ft, and the filter cake may contain up to 50 wt. % solids.

Thickener design criteria have also been developed as a function of the modified fineness modulus. Figure 10 is a plot of thickener unit area requirement as a function of the fineness modulus. As the fineness modulus increases (indicating a smaller particle size distribution) the amount of thickener area required increases dramatically. Figure 10 also includes a correlation for thickener sizing with polymer added. The values shown are for the best settling characteristics that can be achieved by efficient flocculation.

As the particle size distribution decreases the thickener size increases and the percent solids in the thickener underflow drops. So not only is a larger thickener required, but the thickener is able to do less concentrating of the solids. This reduction in underflow concentration is shown in Figure 11 as a function of fineness modulus.

In the past few years the knowledge of how to control particle size distribution in magnesium-enhanced lime scrubbing systems has developed significantly. This knowledge has allowed existing scrubbing systems to operate more efficiently by controlling scrubber operating variables to produce larger particles for subsequent dewatering. However, the improvements are limited by the design of the scrubbing systems.

#### THICKENER AND FILTER SIZING CRITERIA FOR NEW SYSTEMS

New magnesium-enhanced lime scrubbing systems could be designed for larger particle size distributions. The result will be smaller thickeners and filters that produce higher underflow concentrations and drier filter cakes. Both the lab and full-scale results suggest that magnesium-enhanced lime systems be designed for low suspended solids concentrations in the scrubbers, low shear in pumps and agitators, and short residence times to produce larger sludge particles. Assuming that sludge particles can be produced with modified fineness modulus of 1.2 - 1.3, laboratory dewatering data suggest the following criteria for sizing thickeners and filters:

- Full-scale filtration rate ..... 200 - 250 lb/hr/ft<sup>2</sup>
- Filter cake solids concentration ..... 47 - 52 wt. %
- Thickener underflow concentration ..... 38 - 42 wt. %
- Thickener unit area: without polymer ..... 10 - 12 ft<sup>2</sup>/t/d  
with polymers ..... 2 - 4 ft<sup>2</sup>/t/d

These sizing criteria are a dramatic departure from design values used in the past. However, they appear justifiable based on full-scale data on existing systems.

#### DISCLAIMER REQUIRED BY EPA

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.



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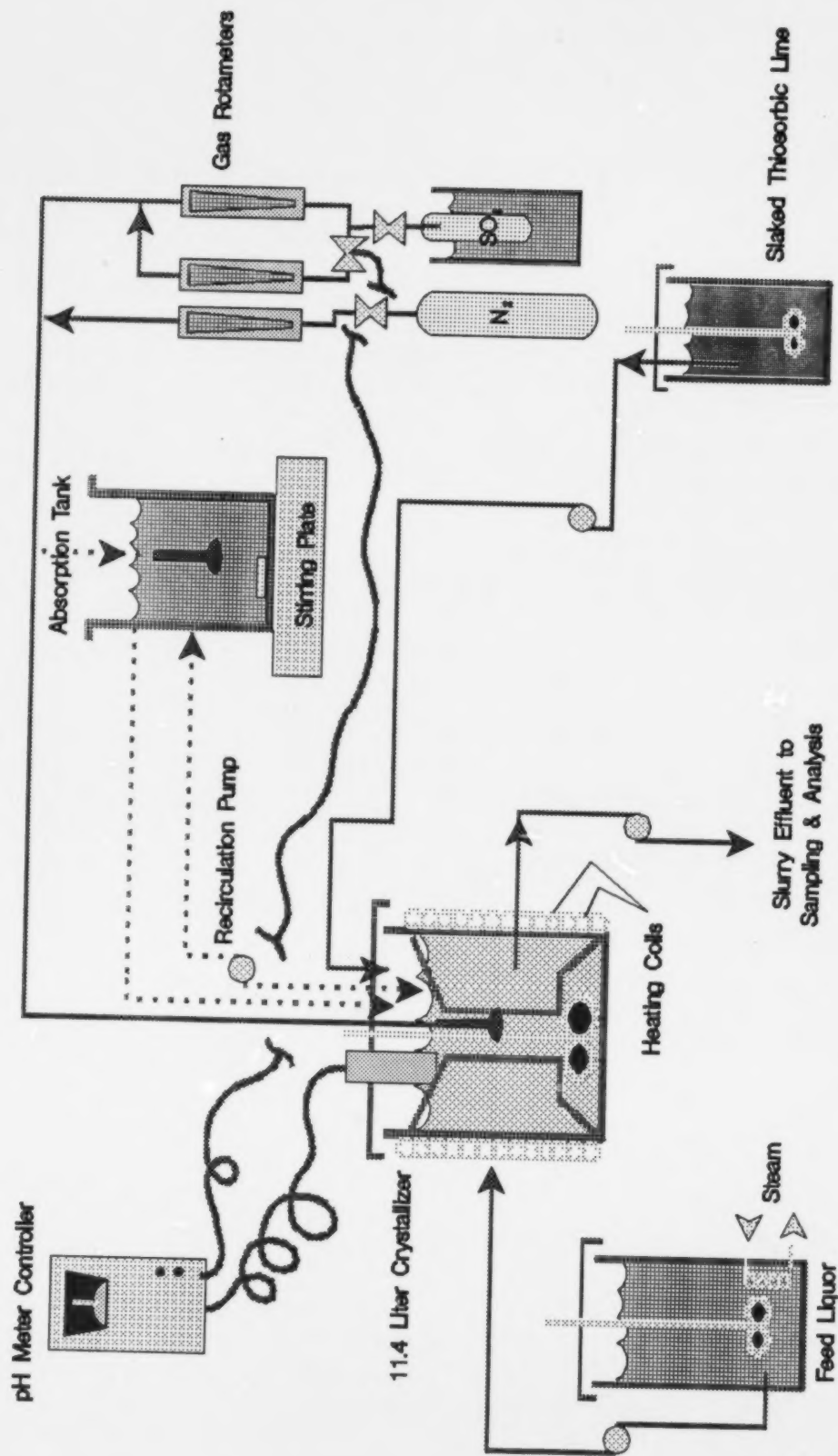


Figure 1. Scrubber simulation apparatus used for the crystallization of  $\text{CaSO}_3 \cdot \frac{1}{2} \text{H}_2\text{O}$ .

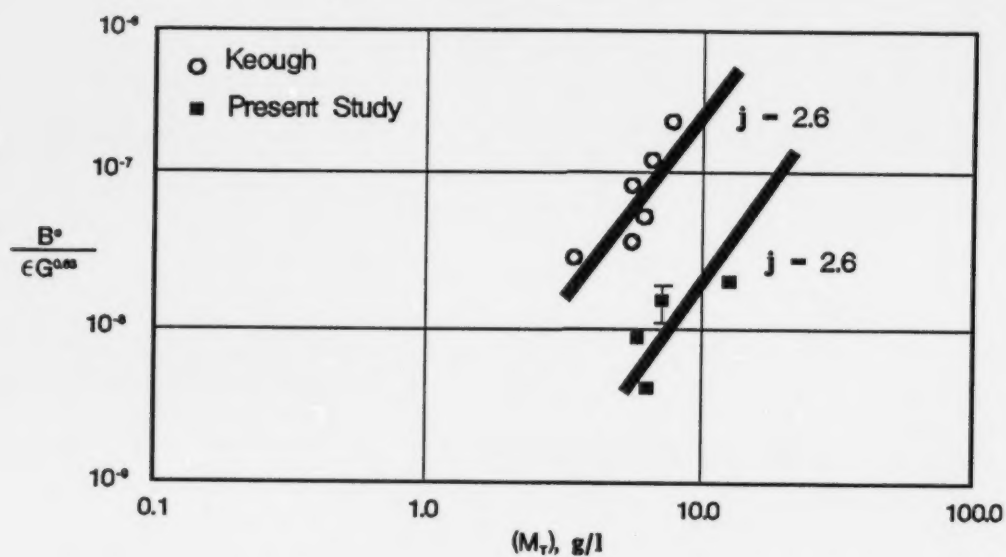


Figure 2: Comparison of data from present study with Keough's nucleation correlation.

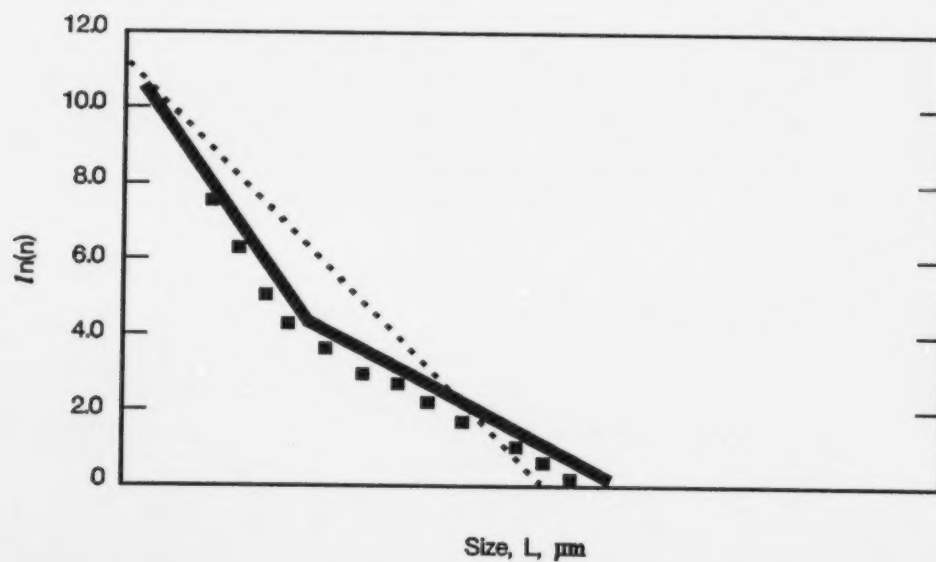


Figure 3. Population density vs. size plot for run 11 with high slurry/absorber recirculation rate.

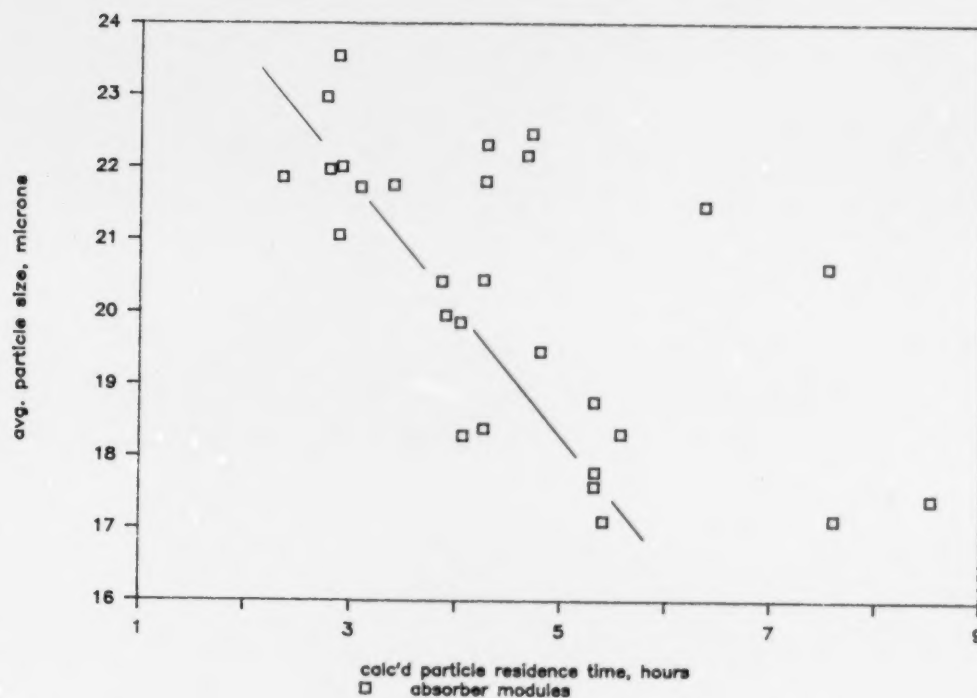


Figure 4. Full-scale dewatering improvement tests, residence time vs. avg. particle size.

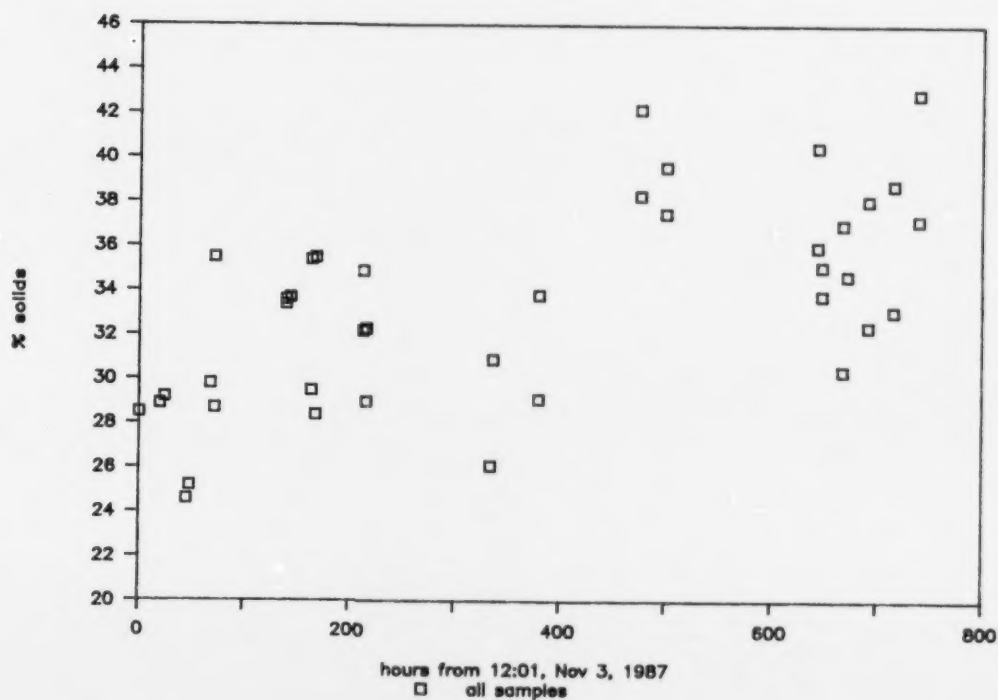


Figure 5. Full-scale dewatering improvement tests, % solids in thickener underflow.



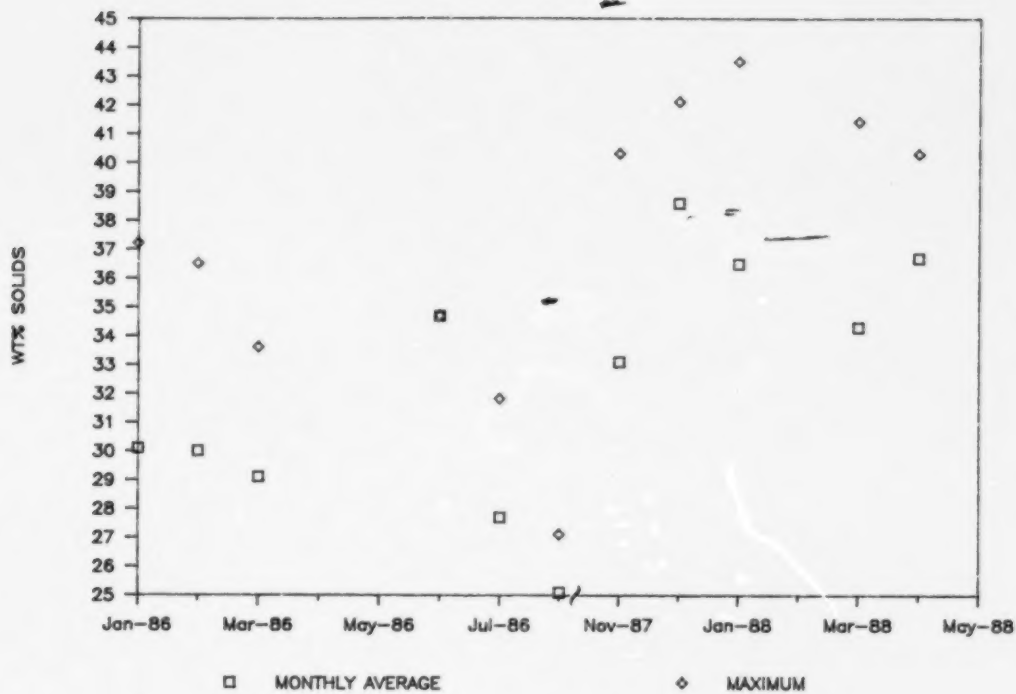


Figure 6. Full-scale dewatering improvement tests, thickener A underflow solids.

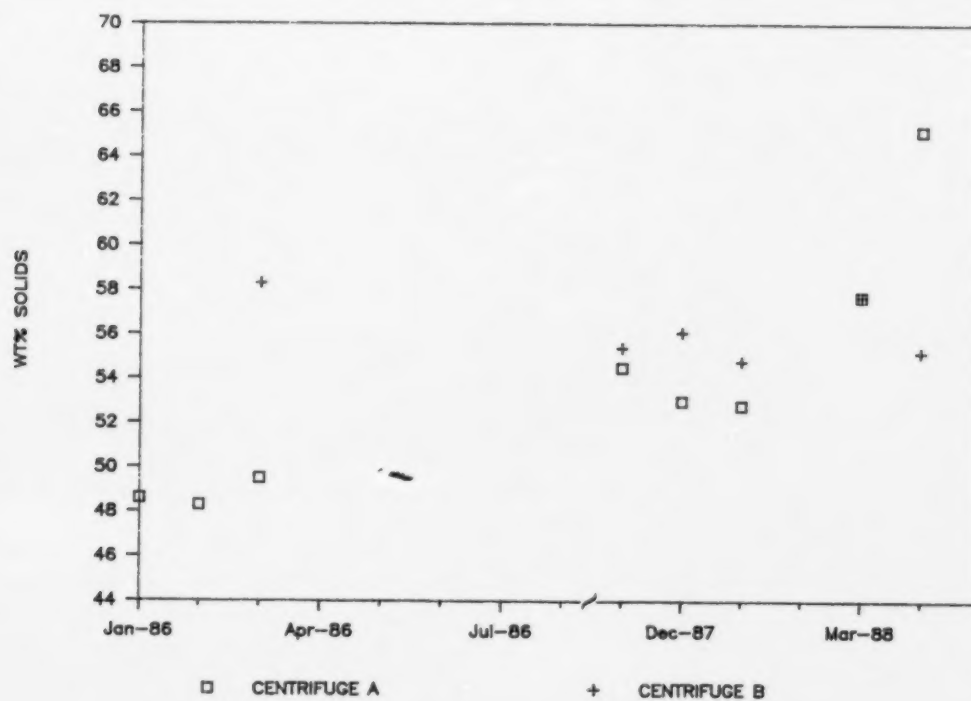


Figure 7. Full-scale dewatering improvement tests, monthly avg. centrifuge cake solids.

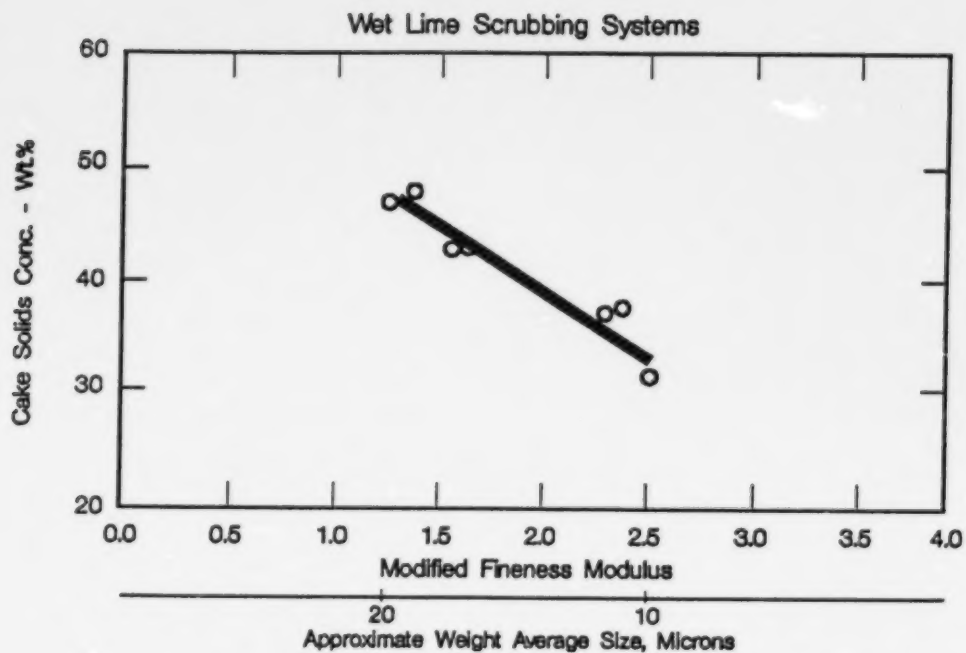


Figure 8. Filter cake solids concentration as a function of modified fineness modulus.

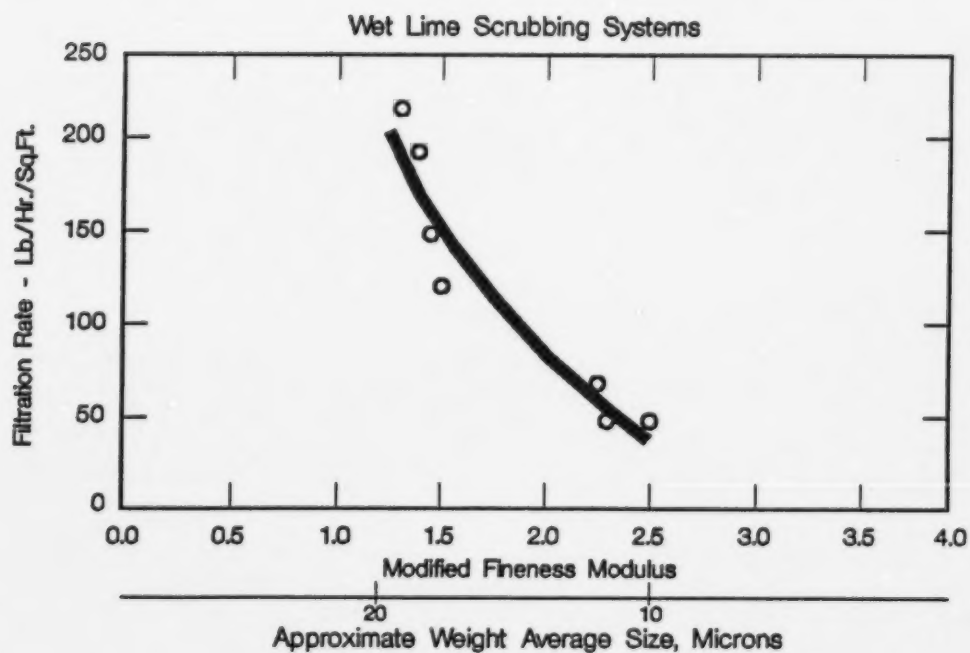


Figure 9. Full-scale filtration rate as a function of modified fineness modulus.

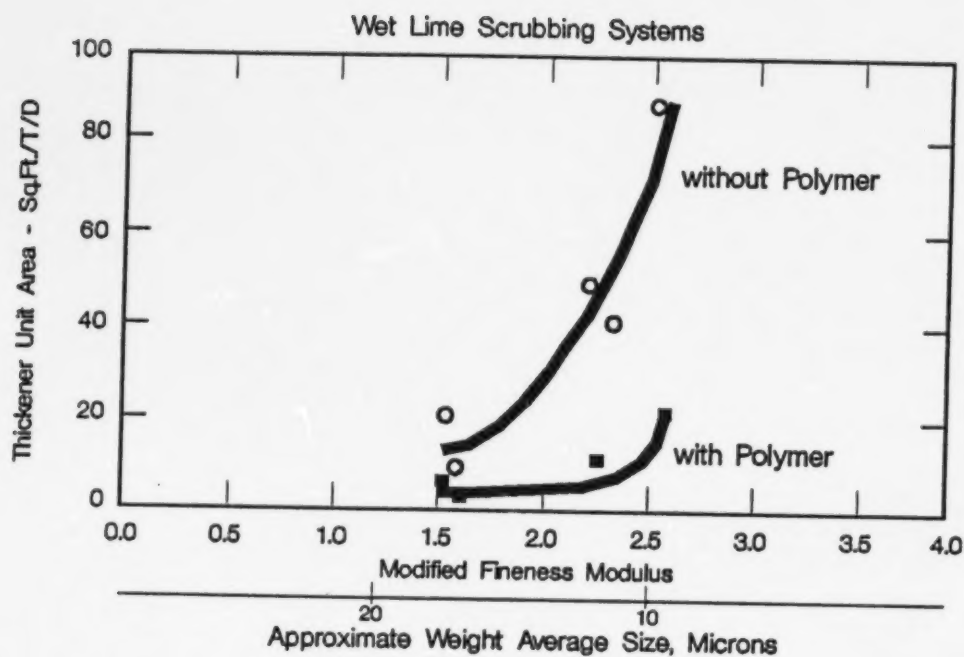


Figure 10. Thickener unit area as a function of modified fineness modulus.

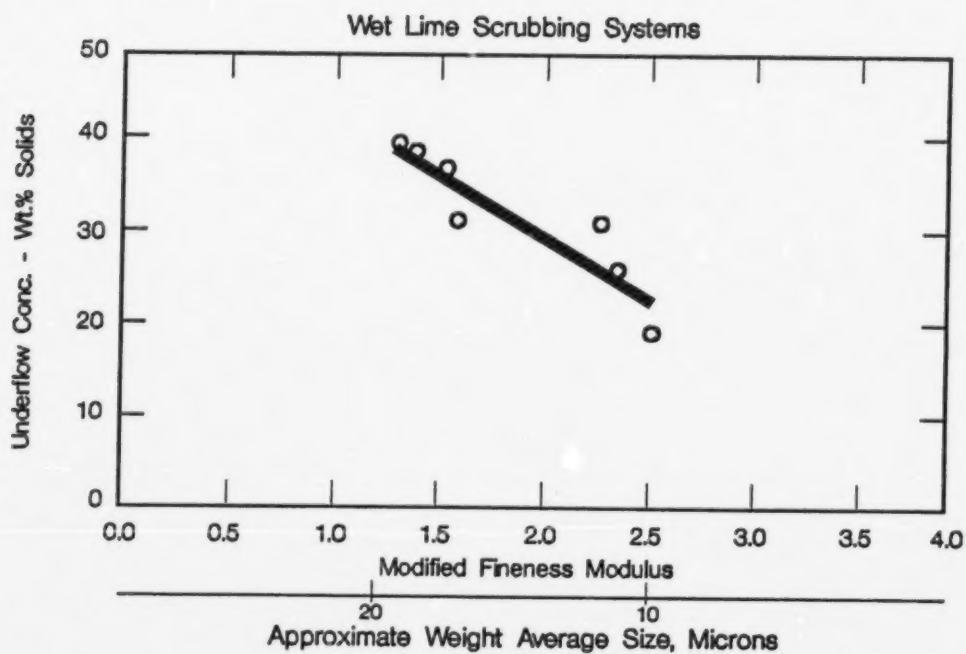


Figure 11. Thickener underflow concentrations as a function of modified fineness modulus.

TABLE 1: BASE RUN CONDITIONS FOR MSMPR

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|                                              |                                                                                             |
|----------------------------------------------|---------------------------------------------------------------------------------------------|
| pH .....                                     | 7                                                                                           |
| Rate of make(a).....<br>(precipitation rate) | 0.125 g $\text{CaSO}_3 \cdot 1/2\text{H}_2\text{O}$ /liter-min                              |
| Residence time(b) .....                      | 60 min                                                                                      |
| Absorption .....                             | In precipitation tank<br>or external absorber                                               |
| Stirrer rpm .....                            | 420 rpm                                                                                     |
| Slurry circulation .....                     | About 2 liters/min of slurry<br>by peristaltic tubing pump;<br>by 3400 rpm centrifugal pump |

---

(a) = set by lime flow

(b) = set by slurry drawdown rate



TABLE 2: OPERATING CONDITIONS

**Feed Solution (Simulated Thickener Return)  
Makeup Concentrations**

|                                                                    |                                                                                 |
|--------------------------------------------------------------------|---------------------------------------------------------------------------------|
| MgCl <sub>2</sub> ·6H <sub>2</sub> O .....                         | 4.3 g/liter = 1500 ppm Cl <sup>-</sup>                                          |
| MgSO <sub>4</sub> ·7H <sub>2</sub> O .....                         | 38.1 g/liter = 14800 ppm SO <sub>4</sub> <sup>-2</sup>                          |
| Mg(OH) <sub>2</sub> .....                                          | 1.82 g/liter                                                                    |
| (Magnesium salts combine to produce Mg concentration of 5000 ppm.) |                                                                                 |
| H <sub>2</sub> SO <sub>3</sub> .....                               | sufficient to provide 1500-2000 ppm sulfite for runs 1-11<br>2500 ppm in Run 12 |
| pH .....                                                           | 7.5-8.5                                                                         |

**Lime Solution Makeup**

 6.9 to 4.4% Ca(OH)<sub>2</sub> solids

**System Operation**

|                                       |                                                                   |
|---------------------------------------|-------------------------------------------------------------------|
| Feed and Crystallizer Temperature.... | 50°C                                                              |
| Crystallizer Stirrer rpm .....        | 585, 685, 1350                                                    |
| Rate of Make (theoretical) .....      | 0.125, 0.25 g/liter-min<br>CaSO <sub>3</sub> *1/2H <sub>2</sub> O |
| SO <sub>2</sub> Temperature .....     | 65°C                                                              |
| Crystallizer pH .....                 | 6.0, 7.0                                                          |
| Absorber Tank Circulation.....        | 2.0 lit/min                                                       |
| Lime Stirrer rpm .....                | 420                                                               |
| Residence Time.....                   | 30, 60, 120 min                                                   |

TABLE 3: COMPARISON OF EXPERIMENTAL AND THEORETICAL CRYSTAL GROWTH FROM RUNS 11 AND 12

| Run | S    | $\sigma$ | G <sub>exp</sub> | G <sub>calc.</sub> |
|-----|------|----------|------------------|--------------------|
| 11  | 3.36 | 2.36     | 0.093            | 0.082              |
| 12  | 3.92 | 2.92     | 0.14             | 0.14               |

S = relative saturation

 $\sigma = S-1$

RECYCLE/REUSE OF BOILER CHEMICAL CLEANING WASTES IN WET LIMESTONE  
FLUE GAS DESULFURIZATION (FGD) SYSTEMS

by: Miriam Stohs and David R. Owens  
Radian Corporation  
Austin, Texas 78720-1088

and

Thomas A. Lott  
Electric Power Research Institute  
Palo Alto, California 94304

ABSTRACT

Boiler chemical cleaning wastes (BCCW) are generated by the periodic water-side cleaning of utility boilers. Treatment of this waste to NPDES discharge standards for iron and copper can be difficult for certain types of BCCW. This paper discusses the results from a series of laboratory tests to determine the feasibility of adding BCCW to a wet limestone flue gas desulfurization (FGD) system. Five different types of BCCW were added to the reaction tank of a bench-scale wet limestone FGD system. Slurry samples were collected at regular intervals, and process performance parameters were monitored continuously. With the exception of one type of BCCW, ammoniated EDTA, metals added with the BCCW precipitated with the scrubber solids. Although both favorable and unfavorable effects on FGD system operation and performance resulted from BCCW addition, all effects were temporary and do not warrant exclusion of this waste management alternative. The addition of two types of BCCW (hydroxyacetic/formic acid and ammoniated citrate) resulted in enhanced  $\text{SO}_2$  removal. The addition of acidic BCCW (HCl and hydroxyacetic/formic acid) resulted in a temporary decrease in limestone utilization. Both ammoniated citrate and ammoniated bromate BCCW also caused temporary decreases in limestone utilization. Sulfite oxidation decreased by 50% or more when ammoniated EDTA, HAF, and HCl BCCW were added.

## INTRODUCTION

Boiler chemical cleaning wastes (BCCW) are generated by the water-side cleaning of utility boilers. Boilers must be periodically cleaned to remove metallic deposits which accumulate on the boiler tube surfaces. Depending on the metals composition of the boiler tubes, BCCW generally contain high concentrations of iron and/or copper, as well as other metals such as chromium, lead, nickel, and zinc.

BCCW treatment and disposal methods include lime precipitation, coponding in an ash pond, evaporation in the fireside of an operating boiler, and contracted off-site disposal. Depending on the type of BCCW, chemical treatment methods achieve varying degrees of success. BCCW which contain chelating agents (EDTA or citric acid) can be especially difficult to treat to National Pollutant Discharge Elimination System (NPDES) limits (1 mg/L for both iron and copper) with conventional lime precipitation.

The Electric Power Research Institute (EPRI) is sponsoring research to evaluate different BCCW treatment and disposal methods. One waste management option under consideration is reuse of BCCW in utility wet flue gas desulfurization (FGD) systems. To investigate this option, a series of laboratory tests were performed in which five different types of BCCW were added to the reaction tank of EPRI's bench-scale wet limestone FGD system. This paper presents the results and conclusions from this study.

## TEST OBJECTIVES

The bench-scale tests were designed to determine the feasibility of adding BCCW to a utility FGD system in terms of BCCW treatment and disposal and FGD system performance and operation. The specific objectives were to determine:

- The fate of the major BCCW components in the FGD system, including metals and cleaning reagents (organic chelating agents and ammonia);
- The effect of BCCW addition on SO<sub>2</sub> removal, limestone utilization, slurry pH, and sulfite oxidation;<sup>2</sup> and
- The relative effects on the leaching characteristics of the waste solids.

## BCCW GENERATION

Boilers typically are cleaned once every 2-5 years, but the cleaning frequency may range from 6 months to 30 years, depending on boiler type and operating history (1). The volume of BCCW produced depends on the boiler tube volume and the number of rinses required to remove all of the solvent. Based on limited data, an average boiler volume correlation is about 125 gallons per electrical megawatt (1). For example, an average 500 MW boiler has a volume of about 62,500 gallons.

Five types of solvents are commonly used for the removal of metal deposits from boiler tube surfaces: hydroxyacetic/formic acid (HAF), ammoniated citric acid, ammoniated ethylenediamine tetraacetic acid (EDTA), ammoniated bromate, and inhibited hydrochloric acid (HCl). Typically, one rinse is required for EDTA,

citrate, HAF, and ammoniated bromate cleaning solutions. Boilers cleaned with HCl-based solutions are usually rinsed at least three times to ensure complete removal of the chloride ion. Based on these values, the average volume of BCCW from one cleaning episode can range from 125,000 to 250,000 gallons for a 500 MW boiler. Since this volume is produced over a period of several hours, flow rates can be on the order of hundreds of gallons per minute.

#### FGD PROCESS DESCRIPTION

Of the existing FGD technologies, scrubbing with a wet lime or limestone slurry has been the most widely applied to utility boilers. In this technology,  $\text{SO}_2$  is absorbed into an aqueous solution of slaked lime,  $\text{Ca}(\text{OH})_2$ , or dissolved limestone,  $\text{CaCO}_3$ , where it reacts to form calcium sulfite and calcium sulfate solids. Waste solids are continuously removed from the system, dewatered, and disposed. Approximately 135 units (representing over 65,000 megawatts of generating capacity) currently use wet lime or limestone FGD systems for  $\text{SO}_2$  removal from flue gas (2).

#### PHYSICAL DESCRIPTION

A typical wet lime/limestone FGD system consists of one or more gas/liquid contactors (spray tower, tray tower, etc.), one or more reaction tanks, a thickener, and solids dewatering and reagent preparation equipment. The scrubber slurry is recycled from the reaction tanks through the gas/liquid contactors where  $\text{SO}_2$  removal occurs. Slurry is continuously removed from the reaction tanks and sent to a thickener or settling pond where waste solids are concentrated. When a thickener is used, the underflow is pumped to solids dewatering equipment (e.g., filters or centrifuges), and both the thickener overflow and final filtrate are recycled to the system.

Waste streams from an FGD system include a filter cake (typically 50-70% solids by weight) and aqueous blowdown. The volume of aqueous blowdown depends on the plant water balance. Many systems operate as closed loop systems in which aqueous waste is removed only in the wet filter cake. If liquid blowdown is necessary, it is often discharged to a holding/evaporation pond. Otherwise, this stream would require treatment to lower the total dissolved solids (TDS) concentration prior to discharge to surface waters. Waste solids from FGD systems normally are disposed of in settling ponds, landfills, or other selected waste disposal sites. A few systems are designed to produce high purity gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) for use in the manufacture of wallboard.

#### OPERATING AND PERFORMANCE PARAMETERS

In this study, the effects of the BCCW on four critical FGD operating and performance parameters were evaluated. These parameters are the reaction tank slurry pH,  $\text{SO}_2$  removal, limestone utilization, and percent oxidation of sulfite to sulfate.

Normally, the pH of the scrubber slurry is a controlled variable in wet FGD systems (i.e., limestone reagent is added on demand to maintain the pH which would otherwise decrease from  $\text{SO}_2$  absorption). Because limestone consumption increases with increasing pH, the operating pH is normally set at the lowest pH at which  $\text{SO}_2$  removal meets the plant's compliance limits. A decrease in the operating pH will adversely affect  $\text{SO}_2$  removal, while an increase in pH will improve the  $\text{SO}_2$  removal.



Limestone utilization is defined as the percent of limestone added which dissolves and reacts with  $\text{SO}_2$  to form calcium/sulfur solids. (The remainder is lost with the waste solids as undissolved limestone.) It is not possible to achieve 100% utilization because of mass transfer limitations which control the limestone dissolution rate; therefore, some level of undissolved limestone is always present in the slurry. The limestone dissolution rate is primarily dependent on the concentration of undissolved limestone, which is referred to as the limestone loading. In general, a given limestone loading is required to maintain the pH at the operating set point. As the limestone loading increases, utilization decreases because more limestone is lost with the waste solids. Because the consumption of limestone increases with decreasing utilization, limestone utilization is a good economic indicator of efficient FGD system operation; in well-operated FGD systems, utilization normally exceeds 90%.

Under certain operating conditions, the limestone dissolution rate can be inhibited as a result of limestone "blinding." This situation has been attributed to the adsorption of certain species on the active dissolution sites of the solid limestone, preventing or limiting particle dissolution. Species which have been proposed as possible causes of blinding include aluminum fluoride complexes and the sulfite ion (3). When blinding occurs, the pH rapidly decreases, and a large increase in limestone loading is required (thereby increasing the limestone dissolution rate) to maintain the desired level of  $\text{SO}_2$  removal. The increase in loading significantly lowers the limestone utilization until the system returns to normal operating conditions.

Absorbed  $\text{SO}_2$  ( $\text{SO}_3^{=}$ ) is oxidized by  $\text{O}_2$  absorbed from the flue gas. This results in the formation of gypsum as well as anhydrite ( $\text{CaSO}_4$ ). The mechanism for sulfite oxidation in an FGD system is not well understood. Some of the factors that are known to affect oxidation are flue gas oxygen concentration, slurry pH and temperature, sulfite concentration, and the presence of trace metals, such as iron and manganese, which catalyze oxidation. The solids oxidation level can influence the settling and dewatering properties of the waste solids and the tendency for gypsum scale to form. In general, FGD solids properties improve with increasing oxidation. Recent studies have shown, however, that in systems that add sulfite oxidation inhibitors, such as thiosulfate, waste solids with good settling and dewatering properties are produced at solids oxidation levels of less than 10% (4).

#### RATIONALE FOR ADDING BCCW TO FGD SYSTEMS

The recycle/reuse of BCCW in a wet lime or limestone FGD system is a conceptually attractive waste management option for several reasons. In terms of waste treatment potential, most of the metals added with the BCCW should precipitate at typical reaction tank pHs (5.4-6.0) and be codisposed with the scrubber sludge. In addition, the alkalinity provided by the lime or limestone can neutralize acidic BCCW.

Addition of BCCW which contain weak organic acids is potentially beneficial, since similar organic acids are intentionally added to some FGD systems to enhance  $\text{SO}_2$  removal (e.g., adipic acid or DBA). However, since the purpose of the organic acids is to solubilize the metals removed during boiler cleaning, their degradation may be desirable to allow chelated metal ions to precipitate. Organic acids may be removed from solution or degraded as a result of chemical (oxidation) and/or physical (coprecipitation) reactions. In addition, dilution of the BCCW may reduce the strength of some chelating agents, which could also result in the release of chelated metal ions. Since many FGD systems have relatively long

liquid-phase residence times, there should be sufficient time for organic acid degradation and metals precipitation to occur.

While the addition of some types of BCCW to wet lime or limestone FGD systems has the potential to enhance  $\text{SO}_2$  removal, BCCW addition may also adversely affect system performance and operation. For example, adverse effects could result from oxidation or pH transients, including an increase in scaling potential and a decrease in limestone utilization. These adverse effects must be fully considered before this disposal method is used at a full-scale facility.

#### EXPERIMENTAL

In this study, seven laboratory tests were performed using EPRI's bench-scale FGD system. One test established the baseline conditions without BCCW addition. Five tests were performed with five different types of BCCW: hydroxyacetic/formic acid, ammoniated citric acid, ammoniated EDTA, ammoniated bromate, and inhibited HCl. One BCCW test was repeated to verify the reproducibility of the results. Undiluted BCCW samples used in the tests were collected at five different power plants as they were drained from the boiler. The results from each BCCW addition test were compared to the baseline test results to determine the relative effects of BCCW addition on FGD system operation and performance.

A schematic of the experimental apparatus is presented in Figure 1. The scrubbing liquor was pumped from the 19-liter reaction tank to a bubbler absorber. In this mode, the contactor simulated the operation of a tray tower. Effluent from the column drained by gravity back into the reaction tank. The simulated flue gas was saturated and heated to produce a typical FGD adiabatic saturation temperature in the bubbler. Limestone reagent was added on demand to maintain a constant pH in the reaction tank. A constant reaction tank temperature was maintained by continuously recirculating the scrubbing liquor through a heated water bath.

The set points of the independently controlled operating parameters are presented in Table 1. To the extent possible, these conditions were selected to simulate a typical full-scale wet limestone FGD system under natural oxidation conditions. The composition of the scrubber slurry at the start of each test simulated a typical wet limestone FGD slurry at a utility firing a medium-sulfur coal, including 3 g/L of fly ash which is absorbed from the flue gas in full-scale systems. (It should be noted that the system was cleaned and fresh solution used for each test.) Makeup water was added as required to maintain a constant system volume of 16 liters in each test.

The duration of each test was approximately 16 hours. Stable conditions were established approximately 2 hours after startup, at which time BCCW addition began. BCCW was added to the reaction tank at a continuous rate of about 6 mL/min over a 2-hour period to simulate the direct draining of BCCW from the boiler to the FGD system. This mode of waste handling would be desirable in a full-scale application, because it requires no BCCW storage. A total of 800 mL of BCCW were added for a dilution factor of 1:20. This would be the approximate dilution factor if 62,500 gallons of undiluted BCCW from a 500 MW boiler cleaning were added to an FGD system with a reaction tank capacity of 1.2 million gallons. After BCCW addition, slurry samples were collected at 1.5-hour intervals, and system operating parameters were monitored continuously.

Due to the small size of the bench scale system, the laboratory test results cannot be scaled-up to predict the actual magnitude of BCCW effects in full-scale

systems. However, the bench-scale system accurately simulates the trends in  $\text{SO}_2$  absorption, the associated liquid phase chemistry, solids precipitation, and limestone dissolution in the reaction tank, which are applicable to full-scale systems. For example, an increase in  $\text{SO}_2$  removal upon BCCW addition to the bench-scale system would indicate that a similar increase would occur in a full-scale system, although the actual magnitude of the increase would not be the same. Therefore, BCCW addition tests in a pilot or full-scale FGD system would be necessary to predict the actual magnitude of BCCW effects in full-scale systems.

## BENCH-SCALE TEST RESULTS

### FATE OF BCCW COMPONENTS

Table 2 summarizes the fates of the primary BCCW components in the scrubber slurry for each bench-scale test. The last two columns present the percent reduction in liquid-phase concentration for each species during the 2-hour waste addition period and over the entire test. (Because of the analytical error associated with the concentration data, only one significant figure is reported for the calculated values in these two columns.) Because the uncertainty of most of the analytical data is  $\pm 10\%$ , a reduction of 10% or less cannot be interpreted as a clear indication that a significant amount of the BCCW component was removed during the test.

#### Hydroxyacetic/Formic Acid

Chromium and iron were both present in this BCCW at concentrations greater than 20 mg/L. (Trace quantities of other metals were present in this waste, but only those components which gave a predicted initial concentration of  $\geq 1$  mg/L are presented in Table 2.) The data presented in Table 2 indicate that both of these metals rapidly disappeared from solution when the waste was added. The organic acids appeared to gradually disappear from solution, with a twenty percent reduction in the post-injection concentration of each species over the course of the test. Possible mechanisms for the removal of these organic acids include oxidative decarboxylation and/or coprecipitation with calcium sulfite solids. [These two mechanisms have been shown to be partially responsible for the disappearance of formic acid in previous bench-scale tests (5).] Because several hundred mg/L of both hydroxyacetic and formic acids were present in solution throughout the test, it can be concluded that the iron and chromium were not strongly chelated by these species.

#### Ammoniated Citric Acid

The concentrations of copper, iron, nickel, and zinc all exceeded 20 mg/L in this waste. The data in Table 2 show that the results of the duplicate citrate tests are consistent. The overall percent reduction in both the copper and zinc was 90% or greater in both tests, while the overall percent reduction in iron and nickel were 70% and 20%, respectively. In both tests the measured post-injection concentration of iron was 50-60% lower than the predicted initial concentration of this element, which indicates that a significant quantity of iron precipitated as the waste was added to the reaction tank. While this does not appear to be the case for nickel, it should be noted that the predicted initial concentration of this species was much lower than that of iron. Because 10-20% of the citrate added with the waste was still in solution at the end of the test, it can be concluded that a percentage of the iron and nickel added with the BCCW remained in solution chelated to the citrate ion. This conclusion is supported by the data



presented in Figure 2 which shows the iron and citrate concentrations measured during the test.

Ammonia was present at a high concentration in this BCCW, resulting in a high ammonia concentration in the scrubber slurry when this waste was added. It appears that this species remained in solution throughout the test. This is not unexpected, since ammonia would be converted to the ammonium ion ( $\text{NH}_4^+$ ) at the slurry pH of 5.7, and would therefore not be lost as gaseous  $\text{NH}_3$ . The conversion of ammonia to ammonium also resulted in the rapid precipitation of copper, which was previously solubilized in the BCCW by complexation with ammonia as  $\text{Cu}(\text{NH}_3)_4^{++}$ . The rapid decrease in copper concentration during this test is illustrated in Figure 3.

#### Ammoniated EDTA

Copper, iron, nickel, and zinc were also present in this BCCW at concentrations greater than 20 mg/L. As can be seen in Table 2, the measured post-injection concentrations of these species were very close to the predicted initial concentrations. This indicates that only a very small amount, if any, of the BCCW metals precipitated during waste addition, which is contrary to the results for the HAF and citrate wastes. Furthermore, the low overall percent reductions of these metals indicate that, at most, only very small amounts precipitated from solution during the rest of the test. Because the EDTA also did not decrease in concentration throughout the test, it appears that the metals remained in solution because they were chelated by EDTA.

This BCCW also contained a high concentration of ammonia. As was observed in the citrate test, a significant amount of ammonia was not removed from solution during the test.

#### Ammoniated Bromate

Both of the metals present in the ammoniated bromate waste, copper and zinc, rapidly disappeared from solution when this BCCW was added to the limestone slurry. This result was not unexpected for copper, since this metal was solubilized by ammonia in the BCCW which was converted to ammonium when the waste was added to the limestone slurry. (It is likely that zinc was solubilized by ammonia as well.) As observed in the other tests with ammoniated BCCW, virtually all of the ammonia remained in solution throughout the test.

#### Inhibited HCl

The same metals present in the citrate and EDTA BCCW - copper, iron, nickel, and zinc - were also present in the HCl BCCW at concentrations exceeding 20 mg/L. Nearly all of the iron and zinc precipitated from solution during the test, while 50% of the copper and 70% of the nickel remained in solution. The rapid decrease in iron concentration is shown in Figure 4. The decrease in the copper and nickel concentrations during the test appears to be dependent on the disappearance rate of thiourea. These data are presented in Figure 5 for copper. It is logical to assume that these metals would completely precipitate from solution when all of the thiourea is consumed, e.g., by oxidative degradation or coprecipitation mechanisms.

### EFFECTS OF BCCW ADDITION ON FGD SYSTEM OPERATION AND PERFORMANCE

Table 3 presents a summary of the effects of BCCW addition on  $\text{SO}_2$  removal, limestone utilization, and sulfite oxidation. The data from each test were



compared to the baseline test results to determine the relative effects of BCCW addition on these parameters. In the baseline test,  $\text{SO}_2$  removal was 74%, at a limestone utilization of greater than 95%, and an average sulfite oxidation of 43%. Addition of the BCCW had several effects on FGD system operation; however, these effects were only temporary. In a full-scale system, the effects of BCCW addition would also be temporary, because the BCCW would be gradually purged from the system.

#### Hydroxyacetic/Formic Acid

As can be seen in Table 3, addition of HAF BCCW enhanced  $\text{SO}_2$  removal.  $\text{SO}_2$  removal enhancement resulting from addition of formic acid has been previously<sup>2</sup> discussed in the literature (5,6). Although the use of hydroxyacetic acid as a buffer additive to FGD systems has not been documented, the acidity constant ( $\text{pK}_a$ ) of this acid is similar to other organic acids which have been used to enhance  $\text{SO}_2$  removal. Addition of this BCCW reduced limestone utilization, because the limestone loading had to be increased to neutralize the waste. Immediately after waste addition, the excess limestone began to dissolve, causing the pH to increase above the operating set point (to pH 5.97). The pH gradually decreased as the limestone loading returned to the pre-injection level. The addition of this waste also significantly reduced the sulfite oxidation. This effect has been observed in previous laboratory tests, although the reason for the reduction in oxidation in the presence of formic acid is unclear (5).

#### Ammoniated Citrate

Addition of this BCCW also enhanced  $\text{SO}_2$  removal because of the buffering properties of citric acid. In spite of the<sup>2</sup> favorable enhancement of  $\text{SO}_2$  removal, citric acid is not used as a buffer additive in full-scale systems because of unfavorable economics. In both citrate tests, the pH initially increased to 5.9 (from 5.7) before rapidly decreasing to 5.51 (first test) and 5.58 (repeat). Excess limestone was added to maintain the pH during waste addition, resulting in an increase in limestone loading. This effect was unexpected because the pH of this waste was 10.3. Apparently, limestone dissolution was inhibited by one or more components in the BCCW, resulting in the rapid decrease in pH and the large increase in limestone loading. This effect may have been caused by the fivefold increase in sulfite concentration which occurred immediately following waste addition, since a high sulfite concentration has been proposed as a possible cause of limestone blinding. The large increase in sulfite concentration is attributable to the high concentration of ammonia resulting from addition of this waste. The system recovered from the blinding condition shortly after waste addition, but dissolution of the excess limestone resulted in a temporary increase in pH (to 5.95 in the first test and 5.75 in the second test) until the pre-injection loading was reached. In spite of the large increase in sulfite concentration, there was no measurable change in sulfite oxidation level as a result of adding this waste.

#### Ammoniated EDTA

Addition of EDTA did not appear to affect  $\text{SO}_2$  removal, nor was an effect on limestone utilization observed in this test. The primary effect of this waste on the FGD system was a drastic decrease in sulfite oxidation, as has been observed in previous laboratory tests (7). The EDTA chelates metal oxidation catalysts, rendering them unreactive. One other effect observed during this test was excessive foaming in the reaction tank. This may have been caused by the presence of surfactants in this BCCW. While foaming can cause operating problems in

full-scale systems, it is likely that this effect could be prevented by addition of an anti-foaming agent.

#### Ammoniated Bromate

The  $\text{SO}_2$  removal increased slightly following waste addition; however, this was a result of the increase in limestone loading and was not caused by buffering properties of components in the BCCW. Although the pH of this waste was 10.4, the pH of the scrubber slurry did not increase when this waste was added (as was initially observed with the citrate waste which also had a high pH). Rather, the pH rapidly decreased to 5.52, causing an increase in limestone loading and a corresponding decrease in utilization. Apparently, components in this waste immediately inhibited limestone dissolution. A 57% increase in sulfite concentration may have contributed to temporary limestone blinding. The addition of bromate, an oxidizing agent, may also have contributed to the observed decrease in utilization; a sudden increase in oxidation would cause the pH to decrease, requiring additional limestone. However, the addition of this waste had little effect on the overall sulfite oxidation level in this test. As was observed in the citrate test, the system began to recover from blinding immediately following waste addition and experienced a similar pH excursion (to pH 6) as the excess limestone dissolved, before returning to the pre-injection loading and pH.

#### Inhibited HCl

There was no significant effect on  $\text{SO}_2$  removal when this waste was added. Removal appears to have been slightly enhanced, but this was a result of the large increase in limestone loading required to maintain the pH while this very acidic waste ( $\text{pH} < 2$ ) was added. This increase in loading also decreased utilization. Following waste addition, the excess limestone began to dissolve, causing a pH excursion (to pH 5.98). The pH gradually decreased as the limestone loading returned to the pre-injection level. This period was longer in the HCl test than in the HAF test, because more excess limestone was required to maintain the pH during HCl waste addition. The calcium concentration nearly doubled (from 28 to 53 mM) as a result of the high concentration of chloride ion added with this BCCW ( $\text{Cl}^-$  increased from 61 to 122 mM), which may also have adversely affected utilization. The sulfite oxidation decreased somewhat in this test, probably because the pH increased to 5.98 following BCCW addition and remained above 5.7 for most of the test. (Metal oxidation catalysts are less soluble at higher pHs.) As was observed in the EDTA test, addition of HCl BCCW caused excessive foaming in the reaction tank which may have been caused by the presence of surfactants in this waste.

#### REGULATORY STATUS OF WASTE SOLIDS

A full review of the regulatory implications of adding BCCW to wet FGD systems was not conducted. However, because metals added with the BCCW may precipitate in a full-scale system, the relative effects on the disposal characteristics of the waste solids in terms of their classification under the Resource Conservation and Recovery Act (RCRA) were examined. To identify potential environmental concerns related to solids disposal, Toxicity Characteristic Leaching Procedure (TCLP) extractions were performed on the slurry solids present at the end of each test. This was a "worst case" scenario, since the solids produced in the bench-scale tests contained higher percentages of precipitated BCCW metals than would be the case if solids had been continuously removed from the reaction tank.

The TCLP results are presented in Table 4. As can be seen, none of the samples exceeded the RCRA toxicity limits. Furthermore, the concentrations for

most of the metals in the TCLP leachates did not differ significantly from the baseline test. Exceptions include slight increases in barium in the leachates from both the HAF and EDTA tests, and a slight increase in chromium for both of the citrate samples. The TCLP results indicate that the addition of BCCW to an FGD system should not alter the RCRA Subtitle C characteristics of the waste solids.

### CONCLUSIONS

Based on the bench-scale results, some of the metals added with the BCCW to an FGD limestone slurry would precipitate from solution and be codisposed with the waste solids. The precipitation of these metals is not expected to adversely affect the RCRA classification of the waste solids. In all tests except for the EDTA BCCW, a large percentage of the BCCW metals disappeared from solution immediately upon addition of the waste to the limestone slurry. However, certain metals added with BCCW containing organic chelating agents remained bound to the organic acids at concentrations exceeding their baseline solubility limits. If conditions in the FGD slurry are not favorable for the degradation of organic acids in the BCCW, chelated metals could remain in solution until they are purged with the aqueous waste. (This could take several months in some full-scale systems.) In addition, if the organic chelating agents do not degrade rapidly, they could solubilize metals normally present at trace concentrations in FGD slurries.

Several conclusions can be made about the effects of BCCW addition on FGD system operation and performance. First, all effects are temporary, with the duration depending on the type of BCCW added and the residence time of the system. Second, the  $\text{SO}_2$  removal should increase or remain the same. No decrease in  $\text{SO}_2$  removal would be expected even if limestone blinding occurred, because the limestone loading could be increased to maintain the  $\text{SO}_2$  removal. The limestone dissolution rate may be inhibited if an ammoniated BCCW is added; however, if blinding occurs the decrease in limestone utilization should only be temporary, and the system should recover shortly after waste addition. The ammonia added would remain in solution as the ammonium ion until being purged with the aqueous waste. Ammonia air emissions are therefore not anticipated to be a problem; however, the effects of high ammonium ion concentrations in the limestone slurry and waste solids should be considered.

The addition of acidic BCCW to the reaction tank results in a higher limestone loading to maintain the pH. This would not adversely affect system operation, except for a temporary decrease in limestone utilization. One method of BCCW addition which might prevent an increase in limestone loading is to add the BCCW directly to the limestone reagent tank where it would be neutralized before entering the reaction tanks. Alternatively, the waste could be adjusted to the slurry pH by in-line neutralization as it is pumped to the reaction tanks. Effects on pH and loading could also be minimized with a slower BCCW addition rate or dilution of the waste with one or more rinses prior to addition.

No increases in sulfite oxidation resulted from the addition of any of the BCCW, while decreases resulted with the EDTA, HAF, and HCl wastes. In general, temporary changes in sulfite oxidation are not favorable in a full-scale system, since the performance of the thickener and dewatering equipment may deteriorate with significant changes in the waste solids composition. Furthermore, if a system were operating supersaturated to gypsum, and subsequently became subsaturated because of a reduced oxidation level, gypsum seed would be lost which could lead to scaling when oxidation returned to the original level. Alternatively, in



systems which add oxidation inhibitors, the addition of BCCW which inhibit oxidation may prove beneficial.

In summary, metals in all of the BCCW except for EDTA precipitated with the scrubber solids. Both favorable and unfavorable effects on FGD system operation and performance resulted when the BCCW were added to the reaction tank. However, the negative effects were not of such a magnitude or duration to preclude this waste management alternative. Finally, analysis of the scrubber solids according to the U.S. EPA TCLP did not identify any metals in excess of RCRA solid waste limits.

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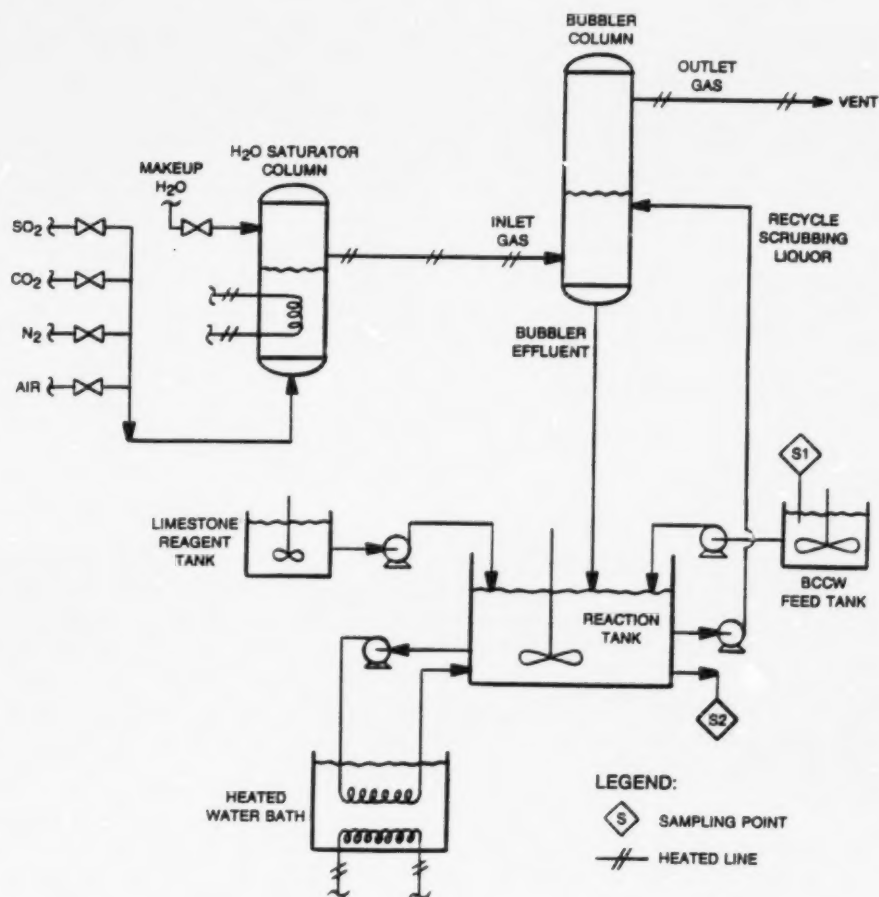


Figure 1. Schematic of EPRI's bench-scale flue gas desulfurization system.

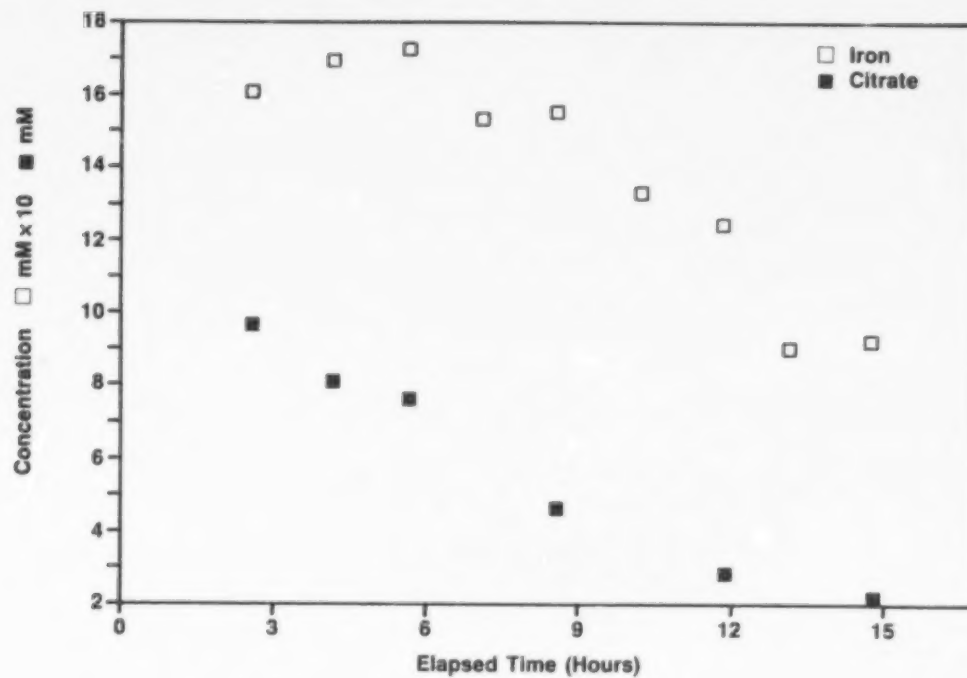


Figure 2. Iron and citrate concentrations during ammoniated citrate BCCW test.

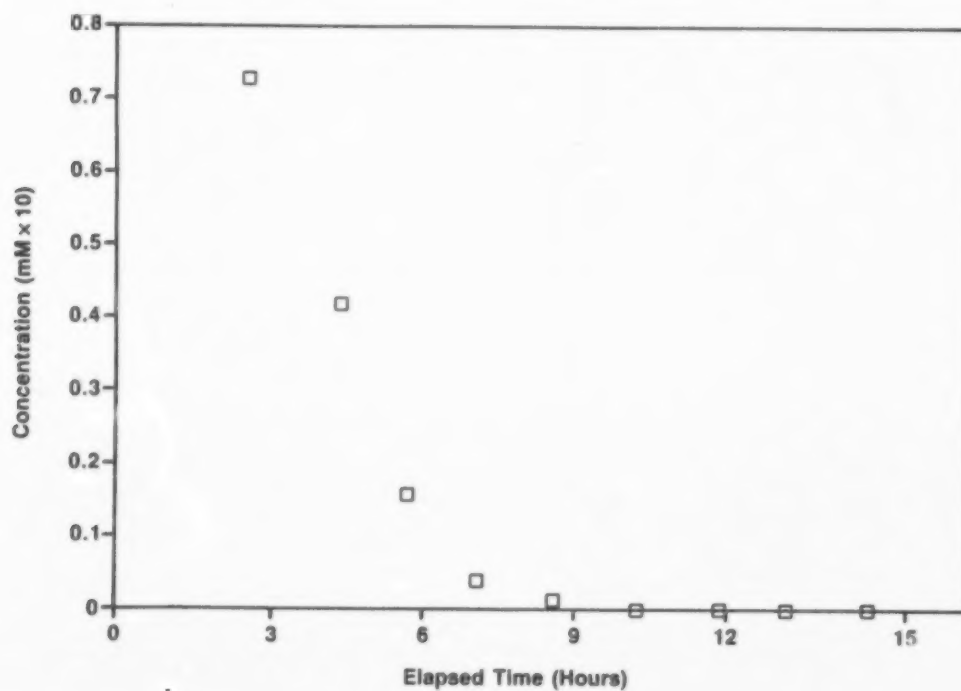


Figure 3. Copper concentration during ammoniated citrate BCCW test.

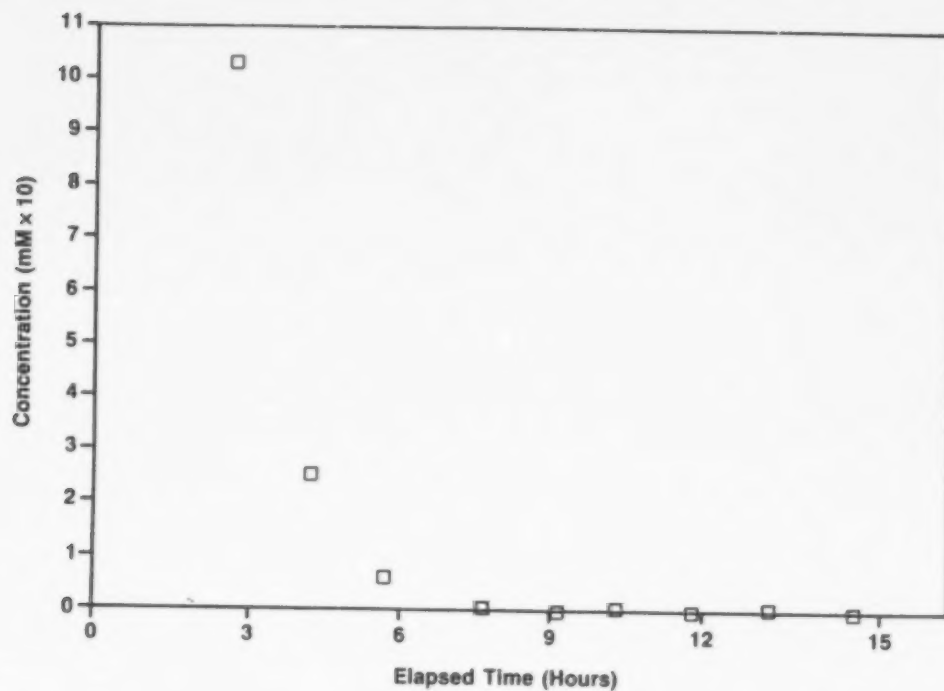


Figure 4. Iron concentration during HCl BCCW test.

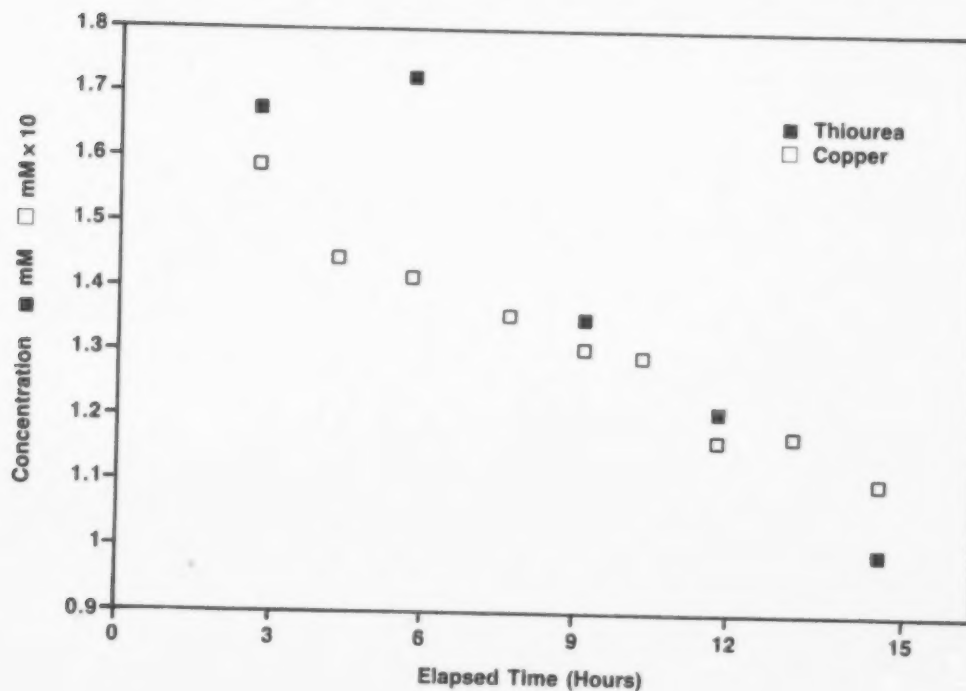


Figure 5. Copper and thiourea concentrations during HCl BCCW test.

TABLE 1. BENCH-SCALE TEST CONDITIONS

| Parameter                                | Operating Set Point |
|------------------------------------------|---------------------|
| Inlet Gas Composition                    |                     |
| Sulfur Dioxide (ppm)                     | 2000                |
| Carbon Dioxide (%)                       | 12.0                |
| Nitrogen (%)                             | 82.8                |
| Oxygen (%)                               | 5.0                 |
| Inlet Gas Flow Rate (SCFM)               | 4.5                 |
| Saturator Temperature (°F)               | 110                 |
| Inlet Gas Temperature (°F)               | 190                 |
| Reaction Tank Temperature (°F)           | 123                 |
| Bubbler Temperature (°F) <sup>a</sup>    | 120                 |
| Reaction Tank pH                         | 5.7                 |
| Recycle Scrubbing Liquor Flow Rate (gpm) | 0.5                 |

<sup>a</sup>Dependent variable.



TABLE 2. FATE OF BCCW COMPONENTS

| Test                           | Waste Component    | Concentration (mg/L) |                                |                             | Percent Reduction During Waste Addition | Percent Reduction Overall <sup>c</sup> |
|--------------------------------|--------------------|----------------------|--------------------------------|-----------------------------|-----------------------------------------|----------------------------------------|
|                                |                    | BCCW                 | Predicted Initial <sup>a</sup> | Post-Injection <sup>b</sup> |                                         |                                        |
| Hydroxyacetic/<br>Formic Acids | Cr                 | 20                   | 1.1                            | ND                          | >99                                     | >99 <sup>d</sup>                       |
|                                | Fe                 | 5,500                | 280                            | 1.2                         | >99                                     | >99 <sup>d</sup>                       |
|                                | Hydroxyacetic Acid | 19,000               | 950                            | 740                         | 20                                      | 40                                     |
|                                | Formic Acid        | 9,500                | 480                            | 420                         | 10                                      | 30                                     |
| Ammoniated Citrate             | Cu                 | 100                  | 5.0                            | 2.9                         | 40                                      | >99 <sup>d</sup>                       |
|                                | Fe                 | 3,900                | 200                            | 81                          | 60                                      | 70                                     |
|                                | Ni                 | 54                   | 2.7                            | 2.5                         | 10                                      | 20                                     |
|                                | Zn                 | 70                   | 3.8                            | 1.4                         | 60                                      | >99 <sup>d</sup>                       |
|                                | Citrate            | 42,000               | 2,100                          | 1,800                       | 20                                      | 90                                     |
|                                | Ammonia (as N)     | 14,000               | 700                            | 1,000                       | --                                      | 10                                     |
| Ammoniated Citrate-repeat      | Cu                 | 100                  | 5.0                            | 4.6                         | 10                                      | >99 <sup>d</sup>                       |
|                                | Fe                 | 3,900                | 200                            | 91                          | 50                                      | 70                                     |
|                                | Ni                 | 54                   | 2.8                            | 2.8                         | <1                                      | 20                                     |
|                                | Zn                 | 70                   | 4.1                            | 2.8                         | 30                                      | 90                                     |
|                                | Citrate            | 42,000               | 2,100                          | 1,900                       | 10                                      | 80                                     |
|                                | Ammonia (as N)     | 14,000               | 700                            | 880                         | --                                      | <1                                     |
| Ammoniated EDTA                | Cu                 | 1,100                | 53                             | 50                          | 10                                      | 10                                     |
|                                | Fe                 | 6,600                | 330                            | 330                         | <1                                      | <1                                     |
|                                | Ni                 | 270                  | 14                             | 13                          | 10                                      | 10                                     |
|                                | Zn                 | 33                   | 2.0                            | 2.0                         | <1                                      | <1                                     |
|                                | EDTA               | 43,000               | 2,200                          | 2,300                       | --                                      | <1                                     |
|                                | Ammonia (as N)     | 7,100                | 360                            | 360                         | --                                      | 10                                     |
| Ammoniated Bromate             | Cu                 | 700                  | 35                             | 17                          | 50                                      | >99 <sup>d</sup>                       |
|                                | Zn                 | 70                   | 3.8                            | 0.45                        | 90                                      | >99 <sup>d</sup>                       |
|                                | Ammonia (as N)     | 8,100                | 410                            | 330                         | --                                      | <1                                     |
| Inhibited HCl                  | Cu                 | 290                  | 15                             | 10                          | 30                                      | 50                                     |
|                                | Fe                 | 4,800                | 240                            | 58                          | 76                                      | >99 <sup>d</sup>                       |
|                                | Ni                 | 270                  | 14                             | 11                          | 20                                      | 30                                     |
|                                | Zn                 | 140                  | 7.5                            | 2.8                         | 60                                      | 90                                     |
|                                | Thiourea           | 4,600                | 230                            | 130                         | 40                                      | 60                                     |

<sup>a</sup>The predicted initial concentration is the BCCW concentration at a 1:20 dilution plus the pre-injection background concentration (in most cases, not detected).

<sup>b</sup>Taken immediately following the 2-hour waste addition period.

<sup>c</sup>Based on the predicted initial and final sample concentrations.

<sup>d</sup>The final concentration was at or below the background concentration.

TABLE 3. EFFECTS OF BCCW ADDITION ON BENCH-SCALE FGD SYSTEM PERFORMANCE

| Test                           | SO <sub>2</sub> Removal (%) | Limestone Utilization <sup>a</sup> (%) | Sulfite Oxidation (%) |
|--------------------------------|-----------------------------|----------------------------------------|-----------------------|
| Baseline                       | 74                          | >95                                    | 43                    |
| Hydroxyacetic/<br>Formic Acids | 92/88                       | 77/>95 <sup>b</sup>                    | 18                    |
| Ammoniated<br>Citrate          | 89/85                       | 66/87                                  | 42                    |
| Ammoniated<br>Citrate-repeat   | 89/84                       | 80/94                                  | 44                    |
| Ammoniated<br>EDTA             | 77/75                       | >95/>95                                | 3                     |
| Ammoniated<br>Bromate          | 82/75                       | 85/>95                                 | 39                    |
| Inhibited<br>HCl               | 82/75                       | 68/>95                                 | 21                    |

<sup>a</sup>Percent of limestone added which reacts with SO<sub>2</sub> to form sulfur solids.

<sup>b</sup>Post-injection/Final.

TABLE 4. CONCENTRATIONS OF METALS IN TCLP LEACHATES OF SLURRY SOLIDS

| Test                           | Concentration, (mg/L) |        |       |        |        |         |       |        |
|--------------------------------|-----------------------|--------|-------|--------|--------|---------|-------|--------|
|                                | Ag                    | As     | Ba    | Cd     | Cr     | Hg      | Pb    | Se     |
| Baseline                       | 0.026                 | <0.002 | 0.47  | <0.003 | 0.023  | 0.0003  | 0.05  | <0.005 |
| Hydroxyacetic/<br>Formic Acids | 0.03                  | <0.002 | 0.77  | <0.003 | 0.021  | <0.0002 | <0.05 | <0.005 |
| Ammoniated<br>Citrate          | 0.03                  | <0.002 | 0.44  | <0.003 | 0.043  | 0.0003  | <0.05 | <0.005 |
| Ammoniated<br>Citrate-repeat   | 0.025                 | <0.02  | 0.17  | 0.007  | 0.047  | <0.0002 | <0.05 | <0.005 |
| Ammoniated<br>EDTA             | 0.02                  | 0.002  | 0.62  | <0.003 | 0.022  | <0.0002 | <0.05 | <0.005 |
| Ammoniated<br>Bromate          | 0.025                 | <0.002 | 0.41  | <0.003 | 0.024  | 0.0002  | <0.05 | <0.005 |
| Inhibited<br>HCl               | 0.03                  | <0.002 | 0.086 | 0.007  | 0.015  | 0.0003  | <0.05 | <0.005 |
| Blank                          | <0.009                | <0.002 | 0.31  | 0.008  | <0.009 | <0.0002 | <0.05 | <0.005 |
| RCRA Limit                     | 5.0                   | 5.0    | 100   | 1.0    | 5.0    | 0.2     | 5.0   | 1.0    |

## DESIGN OF WASTE MANAGEMENT SYSTEMS FOR FURNACE SORBENT INJECTION TECHNOLOGIES

by: Garth D. Guntz  
Mark G. Lees  
Baker/TSA, Inc.  
Coraopolis, Pennsylvania 15108

Dean M. Golden  
Electric Power Research Institute  
Palo Alto, California 94303

### ABSTRACT

This paper presents information concerning the wastes and waste management systems associated with furnace sorbent injection. Waste properties as well as typical waste management systems and their associated costs are considered. Typical waste management systems have been developed based on waste properties, experience at existing generating stations, and regulatory requirements. An overview of the guidance and design information being developed under the EPRI Advanced SO<sub>2</sub> Control Waste Management Project (RP 2708) is provided. Assessment of the the operational and economic impacts of waste management for furnace sorbent injection can be facilitated using this information.

### INTRODUCTION

The Electric Power Research Institute (EPRI) recognizes a need for information concerning the management of wastes produced from advanced SO<sub>2</sub> control technologies. Therefore, the "Advanced SO<sub>2</sub> Control Waste Management Project" (RP 2708) was conceived and funded beginning in 1985. This project, currently in progress, is investigating the management of waste by-products produced by calcium spray drying, atmospheric fluidized bed combustion, lime/limestone furnace injection, sodium alkali injection, and advanced coal cleaning technologies. The work is being performed for EPRI by ICF Northwest (the prime contractor) with subcontract support from Radian Corporation, Baker/TSA, Inc., Battelle Northwest Laboratories, and the University of North Dakota. This paper presents a summary of information developed concerning the management of furnace sorbent injection under this project. Included are descriptions of typical waste properties and waste management systems as well as waste management system costs. That waste management costs are only a fraction of the total costs associated with these clean coal technologies must be recognized; therefore, a comparison of waste management costs should not be used by itself to select an advanced SO<sub>2</sub> technology



for a given plant. Nevertheless, capital and operating costs associated with managing wastes are important factors in evaluating and comparing alternative SO<sub>2</sub> control systems.

## BACKGROUND

Lime/limestone furnace sorbent injection (LFI) technology currently is undergoing full-scale demonstration testing in the United States. At present, five utilities in the United States and Canada have some degree of experience with or are planning to use LFI at six power plants. The first permanent, continuously operating, LFI station in North America should be the Shand Plant located in Saskatchewan, Canada which is planned for start-up in 1992.

LFI technology involves the injection of pulverized calcium-based sorbent such as limestone or hydrated lime into the combustion zone of a furnace above the burners. Testing has demonstrated that SO<sub>2</sub> removal and sorbent utilization are optimized by injection into this upper furnace region. The LFI process produces wastes consisting of combinations of conventional fly ash, unreacted lime and the SO<sub>2</sub> reaction by-products, calcium sulfate and calcium sulfite.

Prior to this study, emphasis has been on optimizing SO<sub>2</sub> removal for the technology with little attention given to LFI waste management. The physical and chemical properties of lime/limestone injection wastes can differ significantly from those of conventional fly ash and wet FGD scrubber sludge. These differences may require significant changes in waste management practices which could have an impact on the overall feasibility and economics of the technology.

## BY-PRODUCT CHARACTERIZATION

Proper design and selection of waste management subsystems requires accurate information regarding both physical and chemical waste properties. For this project, limestone furnace sorbent injection waste samples were obtained from both Ontario Hydro and the Southern Research Institute. Characterization was performed only for samples of fly ash collected from the flue gas. Since sorbent is injected in the upper furnace region, the bottom ash should not be affected significantly by the LFI process. Complete details are provided in EPRI report CS-5783 entitled, Laboratory Characterization of Advanced SO<sub>2</sub> Control By-Products: Furnace Sorbent Injection Wastes (1).

The objective of the LFI waste testing was to determine waste properties so as to represent typical wastes as closely as possible. The testing did not propose to examine extreme operating conditions, nor to bracket all types of possible wastes.

The physical and chemical properties of LFI fly ash are different from those of conventional fly ash and wet scrubber sludge. The differences generally relate to the addition of the calcium-based sorbent for SO<sub>2</sub> removal. LFI by-products contain substantial quantities of anhydrous calcium oxide (CaO) and calcium sulfate (CaSO<sub>4</sub>), which cause exothermic hydration reactions when contacted with water. A typical waste contains about 45 percent fly ash, 25 percent free lime (CaO) and 30 percent calcium sulfate by weight.

## CHEMICAL ANALYSIS

As part of this study, chemical and physical analyses were performed on LFI samples from a full-scale Ontario Hydro operation and a Southern Research Institute pilot test facility. Inductively coupled argon plasma emission spectrometry (ICAP) was used to determine the concentrations of 30 inorganic elements. However, this technique's detection limits are near or above the maximum contaminant limits for public drinking water supplies for five of the elements. Therefore, concentrations of these elements also were measured using atomic adsorption spectrophotometry (AAS). Furthermore, wet chemical procedures were used to analyze for several anionic species. The analytical methods and procedures used are detailed in EPRI report CS-5626, Sampling and Analytic Protocol for Advanced SO<sub>2</sub> Control By-Products (2). A summary of the chemical properties determined for LFI wastes is presented in Table 1.

The elements found in the highest concentrations were aluminum, calcium, iron, magnesium, silicon and sodium which are indicators of fly ash content. High concentrations of silicon (as SiO<sub>2</sub>) indicate that these by-products are potentially pozzolanic. High calcium, and to some extent magnesium, concentrations are direct results of lime addition in the LFI process. Hydroxide fractions typically correspond to concentrations of unreacted lime. High hydroxide levels indicate a potential for significant temperature rises due to hydration reactions if water contacts LFI wastes. In addition, high lime levels in combination with pozzolans provide a potential for cementitious reactions. High alkalinities (reflected in the high calcium, magnesium, and sodium contents) indicate potential additional SO<sub>2</sub> removal capability. The Ontario Hydro LFI waste samples contained elevated concentrations of barium, cobalt, copper, lithium, strontium, titanium, vanadium, lead, and selenium. However, these species did not leach from the wastes under a variety of laboratory conditions.

## PHYSICAL PROPERTIES

As compared to chemical, physical waste properties are more diverse in their meanings and methods of determination. For example, they range from the intuitively obvious, like density, to the relatively obscure, like tackiness, and often are highly specific in their application, as for flowability.

Regarding waste management, physical properties affect storage, transfer, transport, disposal and utilization operations; the first three of these general categories are always of concern while the latter two may be considered alternatives to one another depending on their overall feasibility. Since the focus of this paper is on LFI waste disposal operations, the first four categories are discussed in more detail below; utilization of LFI wastes is considered in another EPRI report: Advanced SO<sub>2</sub> Control By-Product Utilization (EPRI Report No. CS-5076) (3).

### Storage

LFI wastes typically are stored in enclosed vessels such as silos and bins. Most silos and bins are designed with converging section hoppers. Specific flowability tests were conducted on LFI samples from the Ontario Hydro power plant

to assess potential design effects. To simulate a range of probable storage conditions, a series of tests was conducted at two different temperatures and varying storage times. In addition, high humidity conditions, as could be expected during summer, were simulated by preconditioning selected samples with humidified air (85 percent relative humidity at 95°F). Samples at 300°F (assuming instantaneous discharge) were tested both with and without preconditioning. To simulate weekend storage, testing was conducted on samples consolidated for three days at ambient temperature, also prepared with and without preconditioning.

The storage properties of a bulk material under appropriate conditions can be determined by special direct shear, controlled strain tests. Flow properties obtained from these tests include bulk density, effective internal angle of friction during flow, kinetic angle of wall friction (measurement of the adhesion between the wall and the bulk material), and unconfined yield strength. Because consolidating pressures vary within bins (i.e., increase with depth), these flow parameters typically are obtained for increasing degrees of compaction.

To sufficiently characterize flowability, an integration of test results is required. Measurements of bulk density, unconfined compressive strength and kinetic angles of wall friction are plotted as a function of consolidation pressure. Then using a graphical procedure, developed by A. W. Jenike, hopper design criteria can be determined. To estimate the necessary bin parameters requires only a rough estimate of effective angle of internal friction during flow. The effective angle of internal friction during flow was estimated to be approximately 50° for this work. The flowability testing generally indicated that LFI waste is easy to free flowing.

Flowability test results can be used to design hoppers for either mass or funnel flow. Funnel flow occurs if hopper walls are not steep enough to promote uniform flow within the vessel. Material flows by gravity towards the hopper outlet through a channel that forms within a stagnant portion of the material mass. As solids flow through the channel, layers slough off the top of the stagnant mass feeding into the channel. Mass flow occurs when hopper walls are sufficiently steep and smooth.

Mass flow has several advantages over funnel flow for conveying LFI wastes. The "last in - first out" condition in mass flow does not give self-hardening hygroscopic LFI waste any extraordinary opportunity to absorb moisture and harden. Mass flow also provides steady waste flow to the conditioner. This ensures a consistent moisture content and provides for steady discharge of a uniform waste for transport and disposal. In addition, mass flow avoids the added power costs and maintenance activities associated with fluidizers typically used in funnel flow applications. The main disadvantage in using mass flow hoppers is the relatively extreme height requirement that is a result of the steep hopper angles required.

Because of the advantages discussed above, mass flow usually is preferred for most ashes; however, if mass flow design parameters are overly restrictive, funnel flow still may be practical. Flowability test results for LFI waste indicate a combination of these two approaches may be a desirable alternative. Specifically, an expanded flow silo should be considered due to the restrictive mass flow hopper angles and funnel flow outlet diameter. Expanded flow silos have an upper hopper section designed for funnel flow which feeds into a lower hopper section designed for mass flow. Based on the most restrictive design parameters determined for LFI fly ash, an upper funnel flow section would have a minimum outlet diameter of 8.7 feet and the lower mass flow section a minimum outlet diameter of 33.6 inches for reliable flow. The hopper angle required for reliable flow depends on the wall material. LFI fly ash was found to require very steep hopper angles (from



the vertical). For hopper diameters up to 4 feet,  $\theta_c$  values of 12°, 14°, and 19° were determined for the three hopper wall materials, aged carbon steel, high density polyethylene, and 304-2B stainless steel, respectively.

In addition to the expanded flow type silo, a new concept in silo design developed by J. R. Johanson may be suitable. This design approach utilizes a cone-in-cone concept which provides mass flow without the extreme height requirements necessary when restrictive mass hopper angles are required. It is particularly attractive for retrofitting an existing silo. To further evaluate this option, specific testing and data analysis would have to be conducted by J. R. Johanson, Inc.

### Transfer and Transport

Transfer and transport subsystems are similar in that they both involve moving waste from one point to another. Both qualitative and quantitative physical properties affect these subsystems. They not only dictate what type of equipment should be used but also influence its design and operation.

**Qualitative Properties** -- Qualitative properties include: abrasiveness, hygroscopicity, tackiness, corrosivity, and tendency to aerate. Hygroscopic materials will absorb water. They then can become tacky; this is evidenced by solids build-up at the elbows of pneumatic equipment as well as sticking and build-up on mechanical conveyors. Such materials also can exhibit mild corrosive tendencies as evidenced by the high pHs (from 11.8 to 13.0) measured for LFI waste neutral extracts. LFI waste also is easily aerated. This makes it easy to convey pneumatically but also limits the speed with which it can be conveyed on a belt conveyor without being wetted or enclosed.

**Quantitative Properties** -- Quantitative properties that affect transfer and transport include: particle size, bulk density, specific gravity and temperature. A major difference between LFI fly ash and conventional fly ash is particle size. In fact, an average of mass mean diameters (mmd) for conventional fly ash reported in the literature (0.189 mm) was on the order of two to three times greater than the equivalent measure for LFI fly ash samples tested as part of this study (4, 5, 6). The smaller average particle size can be attributed to a greater fine fraction due to the calcium/sulfur reaction by-products. A summary of LFI transfer and transport physical properties is presented on Table 2. The consequences of such properties are more fully discussed in the EPRI report, Lime/Limestone Furnace Injection Waste Management Design Guide.

### Disposal

Physical properties that affect disposal include optimum moisture/maximum density, unconfined compressive strength, and permeability.

**Optimum Moisture Content/Maximum Dry Density** -- Important for sizing and designing waste disposal areas, moisture/density relationships were developed by Radian Corporation as part of this study. Their procedure generally followed ASTM D-698. The optimum moisture contents (OMC) determined as part of this study ranged from 36 percent to 52 percent, averaging 42 percent. Maximum dry densities ranged from 68.1 lb/ft<sup>3</sup> to 80.2 lb/ft<sup>3</sup>, averaging 71.7 lb/ft<sup>3</sup>.



Due to the hydration reactions possible with LFI wastes, the above results require some additional explanation. The reported OMC was based on moisture added to the sample. This includes moisture bound by hydration reactions as well as moisture steamed off (due to the heat generated by the hydration reactions). Therefore, the optimum moisture contents reported reflect the approximate conditioning requirements necessary to bring the waste to OMC but are not true optimum moisture contents. Since, in this work, the reported values for maximum dry density were calculated using the values measured for OMC, they are believed to be lower than the actual in-place densities which would be expected. The maximum dry densities reported were estimated by dividing the wet density (determined by weighing a sample of known volume) by one plus the moisture content reported.

**Unconfined Compressive Strength** -- As part of the study, extensive unconfined compressive strength (UCS) tests were conducted in accordance with ASTM D-1633. The effects of curing time, curing humidity and sample moisture content were examined during these tests. LFI samples from Ontario Hydro were prepared at OMC 5 percent above and 5 percent below using three curing conditions and six curing periods. UCS tests were conducted after one, three, seven, 14, 28 and 56 days curing at 100 percent relative humidity (wet cycle). In addition, UCS tests were conducted on samples cured to 28 and 56 days at a 51 percent relative humidity (air dried) and with a seven day wet/dry cycle.

Although sufficient for design of a conventional landfill, unconfined compressive strengths fluctuated widely making conclusions difficult regarding the effects of curing time, curing humidity and sample moisture content. One fairly consistent trend was a decrease in unconfined strength between 28 and 56 days curing (for over two-thirds of the samples). Also, it does appear that after 56 days of curing, samples prepared at OMC developed the greatest strength followed by samples prepared at OMC plus 5 percent. With respect to curing conditions, no obvious trends were apparent. This may imply that curing conditions, including the wet/dry cycling intended to simulate weathering, do not significantly impact strength.

**Permeability** -- Important in assessing potential leachate generation, permeability is a measure of the rate at which water passes through a material. Typically reported as a coefficient of permeability, the values determined during the study ranged from  $3.8 \times 10^{-9}$  cm/s to  $2.8 \times 10^{-5}$  cm/s, averaging  $6.4 \times 10^{-6}$  cm/s. Literature values ranged from  $5.8 \times 10^{-6}$  cm/s to  $7.35 \times 10^{-6}$  cm/s, averaging  $6.58 \times 10^{-6}$  cm/s (7, 8). For comparison purposes, the permeability designated by the EPA for impermeable liner materials for hazardous waste landfills is on the order of  $1 \times 10^{-7}$  cm/s (typical of clay bases). These low permeabilities indicate that liners may not be necessary for disposal of LFI wastes. Samples were compacted to proctor density and cured for 28 days at 100 percent humidity for all the permeability tests.

#### LEACHATE ANALYSIS

Three types of extract tests were conducted on LFI waste samples: (1) the extraction procedure (EP) toxicity test which is currently required by the federal government to determine whether wastes are hazardous; (2) an ASTM extraction test which more closely resembles actual disposal conditions since a neutral extract solution (distilled water) is used; and (3) a new procedure, the Toxicity Characteristic Leaching Procedure (TCLP), which may replace the aforementioned EP toxicity test.

A comparison of extract analyses to EP toxicity limits is provided in Table 3. As can be seen, the results for all three procedures were below the RCRA maximum concentration limits for the eight elements of concern. Interestingly, when the results of the extract procedures are compared, the TCLP values resemble those for both EP and ASTM tests for barium, cadmium, chromium, mercury, and silver. However, for arsenic, lead and selenium, the TCLP values were on the average about 100 times greater than for the other two.

A summary of the extraction test results is presented on Table 3. Sulfate, nitrate, nitrite, total dissolved solids (TDS) and pH were all above the maximum contaminant limits (MCLs) for drinking water. The data indicate that ASTM concentrations for these constituents are slightly higher than for the EP extracts except for TDS which was consistently higher based on EP testing.

The chemical analyses obtained from the batch extractions may represent actual leachate quality too conservatively. Because LFI waste are alkaline and cementitious, column leachate test results should be more representative of actual leachate quality. Results of column leachate testing conducted by Radian Corporation are presented in EPRI CS-5783 (1).

#### LINER COMPATIBILITY

As part of EPRI RP-2708, Battelle Pacific Northwest Laboratories investigated liner compatibility with LFI waste. Included in the testing program were 20 different synthetic liner materials from different vendors and 10 different admix liners. The synthetic liners were evaluated on the basis of changes in dimensions and weight as well as degradation in strength. For the most part, synthetic liners were tested in accordance with the procedures established by National Sanitation Foundation (NSF) Standard Number 54. To accelerate the test program, Pacific Northwest Laboratories shortened the maximum immersion period from 120 days to 90 days and raised the immersion temperature from 50°C to 70°C. The admix liners were evaluated based on permeability and other physical property changes (5).

Synthetic liners were tested for dimensional change and strength degradation after immersion in a 10 weight percent slurry of LFI waste for one, seven, 30 and 90 days. The results of these tests were evaluated based on criteria developed by the NSF. By these criteria, a liner's physical properties must level off and become stable below maximum limits. A summary of the liner compatibility test results is provided in Table 4.

The most important criterion for an admix liner is the effect of waste leachate on liner permeability. LFI extract was produced by mixing a 3-to-1 ratio of deionized water and LFI waste, respectively. Permeability was measured using a flow-through column in accordance with the ASTM procedure for measuring permeability of granular soils except that leachate flowed up the column to facilitate removal of entrained air pockets in achieving a saturated condition. The waste was compacted using the sliding-weight tamper method (ASTM 1982). Admix liners tested and their permeabilities are presented in Table 5.

Based on the liner compatibility testing, (if a liner is deemed necessary for LFI waste) the best choices appear to be soil-bentonite admixes, asphalt emulsions, compacted clay soils, ethylene vinyl acetate co-polymer, high density polyethylene, and low density polyethylene. However, given the properties of

properly conditioned, placed and compacted LFI wastes, liners often may be redundant and impose an unnecessary additional disposal cost.

## WASTE MANAGEMENT SYSTEM

A waste management system consists of all the equipment and components required to:

- (1) Remove the waste material from the collection device starting at the hopper or particulate collector (the collection devices themselves typically are not included).
- (2) Transfer, store, or condition the waste at the plant site.
- (3) Transport the waste to the ultimate disposal area.
- (4) Place, condition, compact or contain the waste at the disposal area.

Waste management systems consist of the following five principal subsystems: transfer, storage, conditioning, transport, and disposal. The transfer subsystem removes waste material from the collection device or temporary storage facility and moves it to a disposal or recycle silo, or to a temporary storage area. In the storage subsystem, the waste is stored temporarily in silos and bins before being transported to disposal or removed for utilization. After storage the waste may be conditioned to make it more amenable for transport and disposal. The transport subsystem then moves the waste from the plant to the disposal site. Finally the waste is placed in a permanent disposal area.

All five subsystems are interdependent; making a decision regarding one can alter the number of alternatives remaining for the others. Many alternatives also are mutually exclusive; that is, selecting an option for one subsystem may eliminate alternatives for other subsystems. For example, if the waste is conditioned with water, pneumatic conveyors most probably cannot be used for transport. The entire waste management system must be designed and specified as a "best" combination of subsystems based on actual waste properties, pertinent regulations, and utility preference.

## LFI DISPOSAL COSTS

Waste management costs were developed during the project based on a typical electric power plant retrofitted with LFI technology. The design basis was developed based on EPRI's Technical Assessment Guide (TAG), as well as consideration of the type of plant which most likely would use the technology. For example, LFI is considered a true retrofit technology and, therefore, would not be applied to a new plant.

For purposes of developing cost estimates, LFI waste are considered to be transferred via a pressure pneumatic conveyance system to a steel silo for storage. The LFI fly ash then is conditioned to a 32 weight percent moisture content and transported with offroad dump trucks two miles to the disposal site for landfilling. A lined heap landfill was assumed to be required at the disposal



site. Figure 1 provides a process schematic of the waste management system chosen. A summary of major design assumptions is presented on Table 6.

Unit costs of 2.08 mills/kWh or \$20.30/ton were estimated based on the "typical" design basis considered. The leveling factors used assume a 15-year book life, a five-year tax life (due to tax preferences allowed for pollution control equipment), a 12.5 percent discount rate, and a 6 percent apparent escalation rate (10).

A parametric cost sensitivity analysis also was performed. Estimated costs were developed and compared for nine different parameters: plant size, stoichiometric ratio, coal sulfur content, coal ash content, coal heating value, haul road length, transport distance, liner/leachate collection system, and tax life. Retrofit life and tax life were examined for the retrofit plant cases. The cost dependence of each parameter was determined by varying the magnitude of that parameter from its base case value. The results of the parametric cost sensitivity analyses for all the advanced SO<sub>2</sub> control technologies demonstrated that the parameters that had the largest effects on waste quantities generated usually had the greatest impact on waste management costs. Plant size, coal sulfur and ash contents, and coal heating value all had significant effects on waste quantities and, therefore, costs. Over the parametric ranges considered, stoichiometric ratio did not significantly affect either waste quantities or costs. The results for parameters that did not affect waste quantities were variable.

Tax life and haul road length had smaller impacts on costs than did liner use and transport distance. The results of the parametric cost sensitivity analysis for retrofit life showed that costs could be reduced significantly by increasing the life of the plant. The results of these analyses are summarized on Table 7.

#### SIMPLIFIED COST ESTIMATION PROCEDURE

Based upon the results of the parametric cost sensitivity analyses, a simplified cost estimation procedure was developed. This estimation procedure involves simply a plot of expected annual anhydrous waste quantity versus leveled unit cost in dollars per ton of dry anhydrous waste and mills/kWh. This graph, Figure 2, was developed from the costs generated as part of the parametric sensitivity analysis for waste quantity dependent parameters. The costs presented are specific for the waste management system described in the EPRI publication Lime/Limestone Furnace Injection Waste Management Design Guide. A brief summary description of the waste management system is presented on Figure 2.

Estimation of disposal costs for a similar situation requires only estimation of average annual waste quantities. If a 1,000 MW plant is assumed, although it is unlikely that this would be the case for LFI technology, waste management costs can be estimated with a knowledge of the unit's heat rate, average fuel analyses, reagent ratio and reagent purity. The 1,000 MW size was used to facilitate comparisons with other technologies based upon EPRI's Technical Assessment Guidelines. Refer to Lime/Limestone Furnace Injection Waste Management Design Guide for details and an example of the estimation procedure.

Simplified, the procedure consists of estimating fuel consumption in tons/yr at retrofit lifetime load based on unit heat rate, coal heating value, and a 50 percent capacity factor. From fuel consumption, the individual waste generation rate components can be estimated and summed to determine the annual waste quantity. The following waste components must be included:



- Ash based on the ash content of the coal and the fuel consumption.
- Calcium sulfate dihydrate ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) based on coal sulfur fraction and required  $\text{SO}_2$  removal.
- Excess hydrated lime ( $\text{Ca}(\text{OH})_2$ ) based on coal sulfur content and reagent ratio.
- Limestone impurities based on reagent purity, reagent ratio and coal sulfur fraction.

Note that the costs expressed per ton of dry anhydrous waste in Figure 8 are not a function of plant size. Therefore, to estimate unit costs in \$/ton, the unit rating (plant size) needs to be considered only as another variable affecting waste quantity. However, costs expressed per unit of electrical energy are inversely related to unit size and cannot be read directly from Figure 8 for other than a 1,000 MW generating station.

### CONCLUSIONS

Based on the results of EPRI Project 2708, effective disposal management systems can be developed for lime/limestone furnace injection wastes. Although typically finer and more alkaline than many conventional electric utility wastes, such wastes should be manageable without incurring significantly increased costs or operating penalties. Project result highlights include:

- (1) LFI wastes are amenable to disposal using essentially conventional systems, hardware and procedures.
- (2) However, the flowability characteristics of these wastes can present special problems which must be addressed carefully.
- (3) Proper conditioning of LFI wastes can be crucial to successful disposal performance; this also is an area demanding special attention.
- (4) The potential environmental risks posed by these wastes appear minimal.
- (5) If a lined disposal site is required by regulatory authorities, compatible liner materials are available.
- (6) Although not detailed in this paper, LFI by-products also appear to have significant utilization potential.

The work described in this paper was not funded by the U. S. Environmental Protection Agency and, therefore, the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

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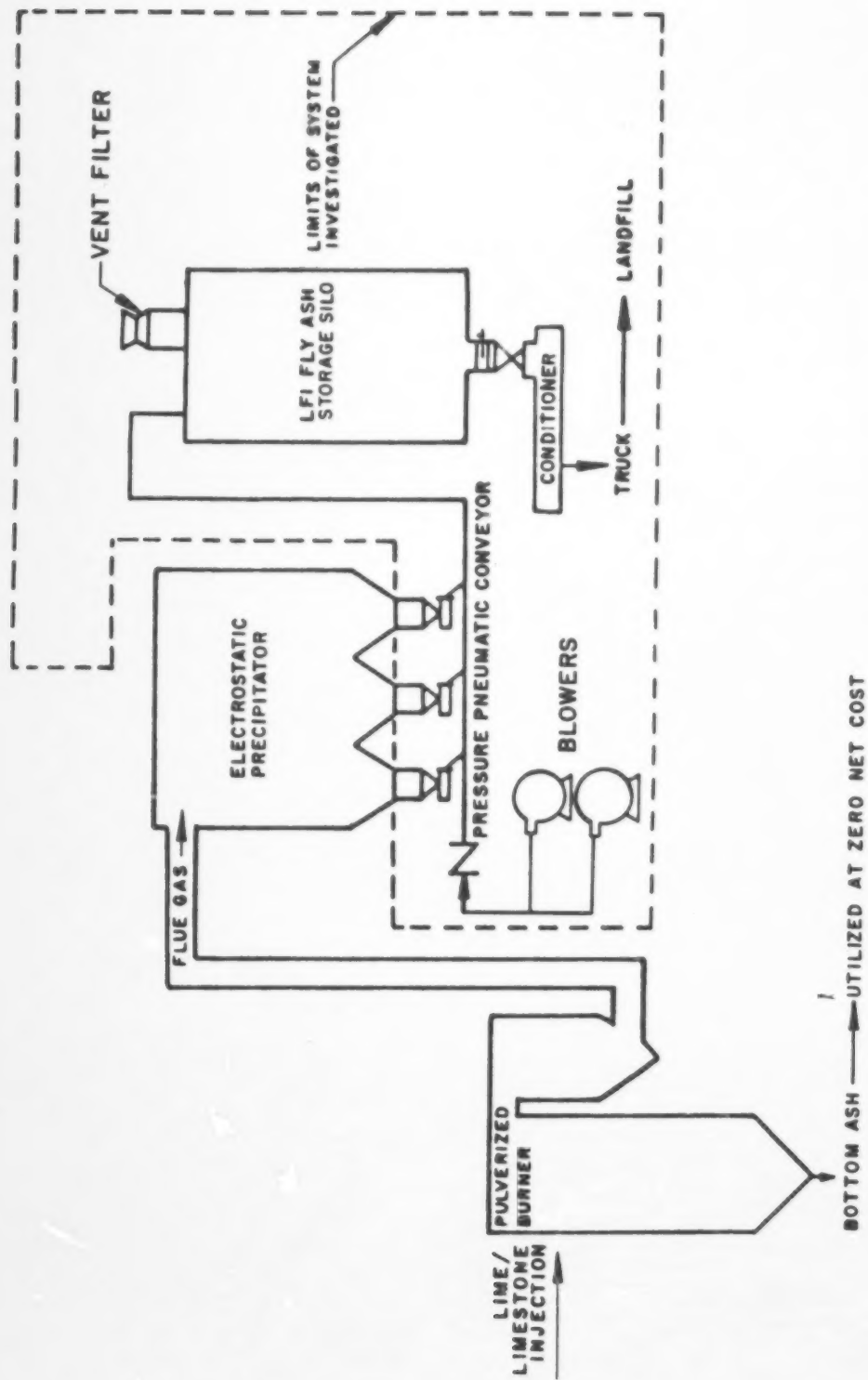


FIGURE 1: LFI WASTE MANAGEMENT PROCESS SCHEMATIC

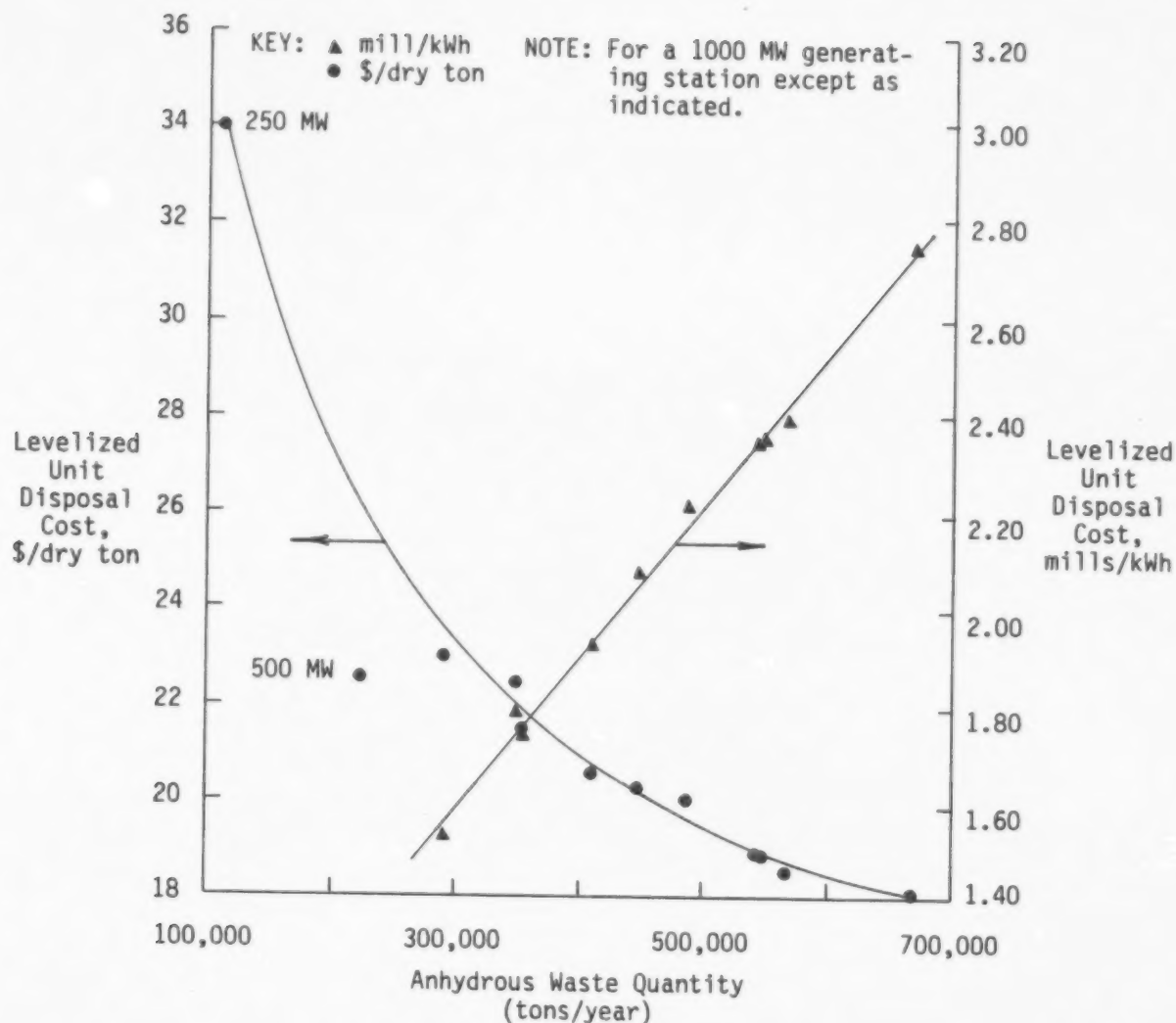


FIGURE 2. LEVELIZED UNIT COST (FROM 1988 TO 2003) AS A FUNCTION OF LFI WASTE QUANTITY

General description of the waste management system concept design for a retrofit limestone furnace injection boiler:

|                           |                                                                                 |
|---------------------------|---------------------------------------------------------------------------------|
| Plant Location:           | Virginia                                                                        |
| Waste Streams Considered: | Fly Ash                                                                         |
| Capacity Factor:          | 50                                                                              |
| Transfer Subsystem:       | Pneumatic pressure conveyance to steel storage silo.                            |
| Transport Subsystem:      | Fly ash is conditioned and transported to landfill using Cat 773-B dump trucks. |
| Disposal Subsystem:       | Fly ash is disposed in a lined, heaped landfill adjacent to a river.            |



TABLE 1. CHEMICAL CHARACTERIZATION OF LFI WASTES  
(mg/kg except where noted)

Inorganic Elements/ICAPES

|                      |                 |
|----------------------|-----------------|
| Aluminum             | 43,500-79,000   |
| Antimony             | <12-24          |
| Arsenic <sup>†</sup> | <12-32*         |
| Barium               | 220-450         |
| Beryllium            | 0.9*-9.0        |
| Boron                | <10-82          |
| Cadmium <sup>†</sup> | <0.4-5          |
| Calcium              | 120,000-285,700 |
| Chromium             | 77-100          |
| Cobalt               | 18-33           |
| Copper               | 22-77           |
| Iron                 | 41,700-59,000   |
| Lead                 | 33*-46*         |
| Lithium              | 24-80           |
| Magnesium            | 3,760-15,000    |
| Manganese            | 140-240         |
| Molybdenum           | 3.3-6.8         |
| Nickel               | 44-110          |
| Potassium            | 4,500-12,300    |
| Selenium             | 67*-230         |
| Silicon              | 73,500-130,000  |
| Silver               | 1.8*-3.5        |
| Sodium               | 1,790-6,560     |
| Strontium            | 180-790         |
| Thallium             | <18             |
| Tin                  | <24-130         |
| Titanium             | 2,460-4,400     |
| Uranium              | <25-87*         |
| Vanadium             | 100-190         |
| Zinc                 | 26-76           |

TABLE 1. CHEMICAL CHARACTERIZATION OF LFI WASTES  
(mg/kg except where noted)  
-continued-

|                                      |             |
|--------------------------------------|-------------|
| Volatile Metals/AA:                  |             |
| Arsenic <sup>+</sup>                 | 15-62       |
| Cadmium <sup>+</sup>                 | <0.1-0.74   |
| Lead <sup>+</sup>                    | 2.4-42      |
| Mercury                              | 0.45-0.93   |
| Selenium <sup>+</sup>                | <0.4-17     |
| Hydroxide (mg/g)                     | 61-198.6    |
| Acid Soluble Sulfur (mg/g as S)      | 20-44       |
| Water Soluble Sulfate<br>(mg/g as S) | 20-44       |
| Water Soluble Sulfite<br>(mg/g as S) | 1.6-47      |
| Water Soluble Chloride (mg/g)        | 0.4*-1      |
| Water Soluble Carbonate (mg/g)       | <2.5        |
| Water Soluble Fluoride (mg/g)        | 0.3-0.4     |
| Radioactivity                        |             |
| Gross alpha (pCi/g)                  | 2.7-28      |
| Gross beta (pCi/g)                   | 26.9-71     |
| Ra-226 (pCi/g)                       | 1.21-3.28   |
| U-235 (pCi/g)                        | 0.0553-0.93 |
| Organics GC & GC-MS As Necessary:    |             |
| Acenaphthene                         | <1          |
| 2, 4-dimethylphenol                  | <2          |
| Fluoranthene                         | <1          |
| Naphthalene                          | <1          |
| Diethylphthalate                     | <1          |
| Dimethylphthalate                    | <1          |
| Benzo (a) pyrene                     | <1          |
| 3, 4-benzofluoranthene               | <1          |
| Chrysene                             | <1          |
| Acenaphthalene                       | <1          |
| 1, 12-benzoperylene                  | <1          |
| Flourene                             | <1          |
| Phenanthrene                         | <1          |
| 1, 2, 5, 6-dibenzanthracene          | <1          |
| Pyrene                               | <1          |

Source: Radian Corporation. Laboratory Characterization of Advanced SO<sub>2</sub> Control Solid Wastes Part 2: Furnace Sorbent Injection Wastes. EPRI RFP 2708. Not yet published.

\* Value is less than 5 times the detection limit.

+ Atomic Absorption provide more accurate volatile metal analysis.

TABLE 2. LFI TRANSFER AND TRANSPORT PHYSICAL PROPERTIES

| Mean Particle Size<br>(mm) |             | Specific Gravity |           | Density (lb/ft <sup>3</sup> ) |           |        |           |
|----------------------------|-------------|------------------|-----------|-------------------------------|-----------|--------|-----------|
|                            |             |                  |           | Poured                        |           | Packed |           |
| Avg.                       | Range       | Avg.             | Range     | Avg.                          | Range     | Avg.   | Range     |
| 0.023                      | 0.018-0.027 | 2.77             | 2.61-3.05 | 44.8                          | 33.2-49.8 | 59.2   | 53.5-69.9 |

TABLE 3. LFI EXTRACT CONCENTRATIONS

| Element                | Leachate Analysis |                |              |
|------------------------|-------------------|----------------|--------------|
|                        | EP                | ASTM           | TCLP         |
|                        | Range             | Range          | Range        |
| As                     | <0.002-0.04       | <0.002-0.004   | 0.25-0.5     |
| Ba                     | 0.2-0.84          | 0.32-0.52      | 0.36-1.0     |
| Cd                     | 0.0003-<0.005     | <0.0005-<0.005 | <0.002-0.013 |
| Cr                     | 0.064-0.18        | 0.083-0.34     | 0.09-0.3     |
| Pb                     | <0.002-0.004      | <0.001-0.008   | 0.12-0.23    |
| Hg                     | <0.0002           | <0.0002        | <0.0002      |
| Se                     | 0.007-0.12        | <0.002-0.049   | 0.16-0.34    |
| Ag                     | 0.015-0.15        | 0.007-0.023    | 0.046-0.088  |
| Cl                     | 18-46             | 54-160         | NA           |
| Cu                     | 0.009-0.026       | 0.004-0.1      | NA           |
| Fe                     | 0.01-0.043        | 0.008-0.11     | NA           |
| Mn                     | 0.001-0.005       | 0.002-0.014    | NA           |
| pH                     | 11.8-12.4         | 11.8-12.5      | NA           |
| SO <sub>4</sub>        | 650-1,600         | 1,300-1,500    | NA           |
| TDS                    | 11,000-13,000     | 4,000-5,400    | NA           |
| Zn                     | <0.003-0.007      | <0.003-0.010   | NA           |
| NO <sub>4</sub> (as N) | 0.4-6.0           | <1-13          | NA           |
| NO <sub>3</sub> (as N) | <1                | <1-6           | NA           |

NA - Not Analyzed.

TABLE 4. LIMESTONE INJECTION WASTE LINER DATA SUMMARY

|     | Liner Material, Vendor                         | Nom.<br>Thick<br>(mil) | Scrim | Change in: |       |      |        |       |
|-----|------------------------------------------------|------------------------|-------|------------|-------|------|--------|-------|
|     |                                                |                        |       | L/Wth      | Thick | Wt.  | Strgth | Elong |
| 1.  | DNA #1952 PVC, Dynamit Noble                   | 40                     | No    | Low        | Mid   | Mid  | High   | Mid   |
| 2.  | DNA #2041 CPE, Dynamit Noble                   | 40                     | No    | High       | High  | High | High   | High  |
| 3.  | DNA #2260L CPE, Dynamit Noble                  | 36                     | Yes   | High       | High  | High | *      | *     |
| 4.  | #M-307 CSPE, Burke Rubber Co.                  | 36                     | Yes   | Low        | High  | High | Mid    | High  |
| 5.  | #M-321 CSPE, Burke Rubber Co.                  | 30                     | No    | High       | High  | High | High   | High  |
| 6.  | PVC, Palco Linings, Inc.                       | 40                     | No    | Low        | Low   | Low  | Mid    | High  |
| 7.  | Dynaloy CPE Alloy, Palco Linings, Inc.         | 30                     | No    | Low        | Mid   | Mid  | Mid    | High  |
| 8.  | Dynaloy CPE Alloy, Palco Linings, Inc.         | 36                     | Yes   | Low        | High  | High | High   | High  |
| 9.  | Dynaloy CPE Alloy, Palco Linings, Inc.         | 45                     | Yes   | Low        | High  | High | High   | High  |
| 10. | HDPE, Gundle Lining Systems                    | 40                     | No    | Low        | Low   | Low  | Mid    | Mid   |
| 11. | Hypalon (CSPE), B. F. Goodrich Co.             | 36                     | Yes   | High       | High  | High | High   | High  |
| 12. | Oil Resistant PVC, B. F. Goodrich Co.          | 30                     | No    | Mid        | High  | High | High   | High  |
| 13. | Low Density Polyethylene, LEC Limited          | 30                     | No    | Low        | Low   | Low  | High   | High  |
| 14. | Ethylene Vinyl Acetate Co-polymer, LEC Limited | 30                     | No    | Low        | Low   | Low  | Low    | Mid   |
| 15. | Ethylene Vinyl Acetate Co-polymer, LEC Limited | 40                     | No    | Low        | Low   | Low  | Mid    | Mid   |
| 16. | Fiberglass Reinforced PVC, National Seal Co.   | 40                     | Yes   | Low        | Mid   | Mid  | High   | High  |
| 17. | Polyester Reinforced PVC, National Seal Co.    | 40                     | Yes   | Low        | Mid   | Mid  | High   | Mid   |
| 18. | Low Density Polyethylene, Poly America, Inc.   | 40                     | No    | Low        | Low   | Low  | +      | +     |
| 19. | XR-5 Ethylene Propylene, Seamans Corp.         | 36                     | Yes   | Low        | High  | High | Mid    | Mid   |
| 20. | Hypalon Industrial Grade, Watersaver Co.       | 36                     | Yes   | Low        | High  | High | High   | Mid   |

## Note:

Low = <5% change at 90 days, 70°C  
 Mid = 5-15% change at 90 days, 70°C  
 High = >15% change at 90 days, 70°C  
 \* = Sample deteriorated - no tensile measurement possible.  
 + = All specimens failed at flaw - no tensile measurement.



TABLE 5. PERMEABILITY RESULTS OF ADMIX LINERS

| Admix Liners                                                                | Permeabilities     |
|-----------------------------------------------------------------------------|--------------------|
| Asphalt Emulsion with Nonwoven Geotextile Fabric                            | $4 \times 10^{-6}$ |
| Hydraulic Asphalt Concrete                                                  | $7 \times 10^{-6}$ |
| Compacted Fine Grain Soils                                                  |                    |
| Exxon Hill Mill, Glenrock, WY<br>(sandy loam: 18% clay, 23% silt, 59% sand) | $2 \times 10^{-5}$ |
| Lucky McMill, Riverton, WY<br>(silty clay: 45% clay, 51% silt, 4% sand)     | $4 \times 10^{-7}$ |
| Shawnee Site, Paducah, KY<br>(silty loam: 20% clay, 74% silt, 6% sand)      | $2 \times 10^{-8}$ |
| Soil/Bentonite                                                              |                    |
| 6% Bentonite                                                                | $3 \times 10^{-7}$ |
| 12% Bentonite                                                               | $5 \times 10^{-8}$ |
| Soil/Cement                                                                 |                    |
| 5% Bentonite                                                                | $2 \times 10^{-5}$ |
| 10% Bentonite                                                               | $2 \times 10^{-4}$ |
| Composite Cover Material                                                    |                    |
| 75% 1.9-cm-minus road base/13% topsoil/4% lime/8% alum                      | $6 \times 10^{-6}$ |

TABLE 6. MAJOR DESIGN ASSUMPTIONS FOR RETROFIT LFI POWER PLANT  
SCENARIOS I and II

| Design Parameter                                 | Assumed Value                            |
|--------------------------------------------------|------------------------------------------|
| Plant Size, MWe                                  | 1000                                     |
| Station Design Life (book life), yrs             | 15                                       |
| Tax Recovery Period, yrs                         | 5                                        |
| Coal Type                                        | Modified<br>Illinois No. 6<br>Bituminous |
| Average Coal Analysis                            |                                          |
| Ash, %                                           | 16                                       |
| Sulfur, %                                        | 2.0                                      |
| Heating Value, Btu/lb                            | 10,100                                   |
| "Worst Case" Coal Analysis                       |                                          |
| Ash, %                                           | 19                                       |
| Sulfur, %                                        | 2.6                                      |
| Heating Value, Btu/lb                            | 9,800                                    |
| Capacity Factor, %                               |                                          |
| Maximum Peak Load                                | 100                                      |
| Average Peak Load                                | 100                                      |
| New Retrofit Unit Load                           | 60                                       |
| Retrofit Lifetime Load                           | 50                                       |
| Stoichiometric Ratio<br>(sorbent: hydrated lime) | 2.0                                      |
| SO <sub>2</sub> Removal, %                       | 50                                       |
| Transport Distance, miles                        | 2                                        |
| Haul Road Length, miles                          | 1.75                                     |
| Degree of Waste Compaction, %*                   | 90                                       |

\* Based on the maximum dry density determined in  
accordance with ASTM D698.

TABLE 7. PARAMETRIC SENSITIVITY ANALYSIS COST SUMMARY  
(December 1987 Dollars)

| Case No | Variable                | Value  | Capital Requirement (x 10 <sup>3</sup> ) |                  |           |       |                       |          |                            |                           |       |          | Operating Cost (x 10 <sup>3</sup> ) |                                    |                                             |            | Levelized Unit Cost * |  |
|---------|-------------------------|--------|------------------------------------------|------------------|-----------|-------|-----------------------|----------|----------------------------|---------------------------|-------|----------|-------------------------------------|------------------------------------|---------------------------------------------|------------|-----------------------|--|
|         |                         |        | Plant                                    | Disposal         |           |       | Impoundment Lost Cost | Indirect | Total Capital Requirement* | Levelized Carrying Charge | Plant | Disposal | Total*                              | Levelized Operating Cost (\$/yr) * | Total Levelized Cost (x 10 <sup>3</sup> ) * | Miliv/ kWh | \$/Ton                |  |
|         |                         |        |                                          | Land Development | Equipment | Land  |                       |          |                            |                           |       |          |                                     |                                    |                                             |            |                       |  |
| 1       | Scenario                | -      | 3,893                                    | 16,171           | 4,531     | 1,332 | 1,315                 | 6,335    | 33,577                     | 6,010                     | 416   | 1,703    | 2,120                               | 3,078                              | 9,088                                       | 20.27      | 2.08                  |  |
| 2       | Plant                   | 500 MW | 1,961                                    | 8,652            | 2,028     | 751   | 658                   | 3,397    | 17,447                     | 3,123                     | 258   | 1,071    | 1,330                               | 1,931                              | 5,054                                       | 22.54      | 2.31                  |  |
| 3       | Size                    | 250 MW | 1,356                                    | 5,795            | 1,693     | 519   | 329                   | 2,390    | 12,082                     | 2,163                     | 205   | 933      | 1,138                               | 1,652                              | 3,815                                       | 34.03      | 3.48                  |  |
| 4       | Stoichiometric          | 1.5    | 3,845                                    | 14,274           | 4,196     | 1,177 | 1,315                 | 6,007    | 30,815                     | 5,516                     | 410   | 1,609    | 2,019                               | 2,931                              | 8,447                                       | 20.57      | 1.93                  |  |
| 5       | Ratio                   | 2.5    | 3,939                                    | 18,667           | 5,443     | 1,548 | 1,315                 | 6,726    | 37,639                     | 6,737                     | 422   | 1,646    | 2,068                               | 3,002                              | 9,739                                       | 20.02      | 2.22                  |  |
| 6       | Sulfur                  | 1.0%   | 3,490                                    | 13,354           | 3,860     | 1,138 | 1,315                 | 5,365    | 28,523                     | 5,106                     | 377   | 1,511    | 1,888                               | 2,741                              | 7,846                                       | 22.47      | 1.79                  |  |
| 7       | Content                 | 3.0%   | 4,294                                    | 19,464           | 5,957     | 1,556 | 1,315                 | 7,376    | 39,962                     | 7,153                     | 455   | 1,741    | 2,197                               | 3,189                              | 10,343                                      | 18.87      | 2.36                  |  |
| 8       | Ash                     | 6%     | 3,420                                    | 11,591           | 2,699     | 983   | 493                   | 5,032    | 24,218                     | 4,335                     | 368   | 1,258    | 1,626                               | 2,361                              | 6,696                                       | 23.00      | 1.53                  |  |
| 9       | Content                 | 10%    | 3,496                                    | 12,747           | 4,416     | 1,053 | 822                   | 5,385    | 27,918                     | 4,997                     | 378   | 1,421    | 1,799                               | 2,612                              | 7,609                                       | 21.49      | 1.74                  |  |
| 10      | Heating                 | 6800   | 4,441                                    | 23,081           | 7,205     | 1,804 | 1,954                 | 8,193    | 46,678                     | 8,355                     | 474   | 2,091    | 2,565                               | 3,724                              | 12,079                                      | 18.09      | 2.76                  |  |
| 11      | Value                   | 8000   | 4,318                                    | 19,845           | 5,957     | 1,587 | 1,661                 | 7,453    | 40,820                     | 7,307                     | 458   | 1,749    | 2,208                               | 3,206                              | 10,512                                      | 18.55      | 2.40                  |  |
| 12      | Haul Road               | 0.25   | 3,893                                    | 15,034           | 3,525     | 1,216 | 1,315                 | 6,035    | 31,018                     | 5,552                     | 416   | 1,426    | 1,842                               | 2,675                              | 8,227                                       | 18.35      | 1.88                  |  |
| 13      | Length                  | 0.75   | 3,893                                    | 15,413           | 3,903     | 1,254 | 1,315                 | 6,126    | 31,904                     | 5,711                     | 416   | 1,373    | 1,789                               | 2,597                              | 8,308                                       | 18.53      | 1.90                  |  |
| 14      | Liner                   | No     | 3,893                                    | 3,068            | 4,531     | 1,332 | 1,315                 | 4,231    | 18,370                     | 3,288                     | 416   | 1,704    | 2,120                               | 3,078                              | 6,366                                       | 14.20      | 1.45                  |  |
| 15      | Tax Life                | 15 yr. | 3,893                                    | 16,171           | 4,531     | 1,332 | 1,315                 | 6,335    | 33,577                     | 6,783                     | 416   | 1,704    | 2,120                               | 3,078                              | 9,861                                       | 21.99      | 2.25                  |  |
| 16      | SO <sub>2</sub> Removal | 70%    | 4,288                                    | 19,304           | 5,957     | 1,548 | 1,315                 | 7,208    | 39,621                     | 7,092                     | 449   | 1,739    | 2,188                               | 3,177                              | 10,269                                      | 18.90      | 2.35                  |  |
| 17      | Transport               | 5 mi.  | 3,893                                    | 16,171           | 2,988     | 1,332 | 1,315                 | 6,327    | 32,026                     | 5,733                     | 416   | 1,973    | 2,389                               | 3,470                              | 9,202                                       | 20.52      | 2.10                  |  |
| 18      | Distance                | 10 mi. | 3,893                                    | 16,171           | 3,558     | 1,332 | 1,315                 | 6,381    | 32,650                     | 5,844                     | 416   | 2,482    | 2,898                               | 4,208                              | 10,052                                      | 22.42      | 2.30                  |  |
| 19      |                         | 20 mi. | 3,893                                    | 16,171           | 4,793     | 1,332 | 1,315                 | 6,496    | 34,000                     | 6,086                     | 416   | 3,570    | 3,906                               | 5,788                              | 11,874                                      | 26.48      | 2.71                  |  |
| 20      | Plant Life              | 10 yr. | 3,893                                    | 11,203           | 3,958     | 859   | 1,315                 | 5,660    | 26,888                     | 5,754                     | 404   | 1,653    | 2,057                               | 2,718                              | 8,472                                       | 20.99      | 2.15                  |  |
| 21      |                         | 20 yr. | 3,893                                    | 20,367           | 4,601     | 1,827 | 1,315                 | 7,029    | 39,032                     | 6,401                     | 428   | 1,753    | 2,181                               | 3,418                              | 9,819                                       | 19.91      | 2.04                  |  |

\* Totals may not add up precisely due to intermediate rounding.  
• Levelizing period 1988 - 2003.

## ENVIRONMENTAL ASSESSMENT OF AN FGD SLUDGE DISPOSAL SITE

by: Ishwar P. Murarka  
Electric Power Research Institute  
Palo Alto, California 94304

Dhanpat Rai and John M. Zachara  
Battelle, Pacific Northwest Laboratories  
Richland, Washington 99352

### ABSTRACT

Sludge produced by the wet FGD technology is generally disposed of in surface impoundments. The chemical composition of these residues suggests that the potential for groundwater pollution should be assessed. In a study sponsored by EPRI (RP2485-08), field measurements were made to assess the leachate composition and the movement of chemicals from a fifteen-year-old FGD sludge pond. Researchers sampled and chemically analyzed the sludge solids and the interstitial waters in the sludge and in the underlying soils. A combination of direct and indirect techniques was used to predict concentration-controlling reactions. Precipitation/dissolution reactions involving solid phases of Ca, S, and several trace elements (Cd, Cu, Ni, Pb, and Zn) were predicted to control leachate concentrations of these elements. The results also show that several other chemicals (e.g., As, B, Ca, F, K, Mg, Mo, Si, sulfur species, and Sr) were found in the leachates formed within the waste unit. However, all of the toxic metals (e.g., As, Cd, Cr, Cu, Ni, Se, and V) remained within the waste unit and showed no migration into the underlying soils or the groundwater. Boron was found to be the best tracer of leachate migration. The boron distribution profile indicated a maximum leachate migration distance of 3 meters and suggested slow leachate migration through and chemical attenuation by the underlying clayey subsoils.



## INTRODUCTION

To protect the nation's air quality, removal of gaseous sulfur species is required by the regulations stemming from the Clean Air Act. Further reductions in sulfur emissions are likely to increase the use of flue gas desulfurization (FGD) technologies in the future. Flue gas desulfurization technologies produce sludges as a result of the use of calcium carbonate, calcium oxide (lime), or alkaline fly ashes to remove (or scrub) gaseous sulfur species from the flue gas stream of a coal-fired electric power plant. Approximately 12.4 million metric tons of FGD sludges were produced in 1986 (1), as compared to only 2 million tons in 1980 (2). The quantities of FGD wastes are expected to reach 50 million metric tons by the end of this century (3). Because 1) large quantities of these residues are disposed of on the land and 2) aqueous concentrations of some of the environmentally important elements are high in sludge leachates, alternative disposal management strategies can be made more effective using long-term predictions of the leaching and migration into underlying soils. The results reported in this paper are from a detailed study (4) of a fifteen-year-old FGD sludge disposal site. This completed research is part of the Electric Power Research Institute's (EPRI) Leaching Chemistry Studies contract (RP2485-08) aimed at developing fundamental information and models to accurately predict leachate generation from field-scale disposal of fossil fuel combustion wastes.

## DESCRIPTION OF THE DISPOSAL SITE

The FGD site is situated in the midwestern United States and receives approximately 63.5-76.2 cm of precipitation annually. The disposal site, located on the flanks of a gradually sloping fluvial basin, occupies an area of approximately 242,760 square meters (60 acres) and was created by constructing a dike with bottom ash to contain the FGD sludge. The FGD sludge was derived from a plant that burned a subbituminous midwestern coal and sent the flue gas through a Venturi scrubber that used  $\text{CaCO}_3$  as the adsorption agent. The sludge was pumped directly into the disposal basin from 1972 to 1979. Subsequently, the pond was retired from active service, and no standing water remained in the basin except during periods of heavy rainfall.

The sludge in the basin is highly stratified in particle size and color. The sludge surface is now partly vegetated. The near-surface zone of the sludge is unsaturated, oxidized, and brownish-gray in color, and it looks like a natural soil. At depths ranging from nearly a meter at the southwest corner of the pond to 0.2 m at the north corner, however, the sludge is saturated, highly reducing chemically, gray to dark gray in color, and thixotropic. The reduced, thixotropic sludge is gel-like in consistency. The presence of highly reducing sludge near the surface attests to the limited diffusion of oxygen through the sludge layers.

## FIELD SAMPLING METHODS AND EXPERIMENTAL PROCEDURES

Cores of the FGD sludge and underlying soils were taken in June 1986 with a hollow-stem rotary auger corer. The drilling rig was equipped with wide tracks to allow it to travel on the soft surface of the sludge. The core materials were collected in 6 x 76 cm Lexan® core barrels. Cores were taken at three locations separated by approximately 150 m across the surface of the sludge pond. At each coring location, pH and Eh were measured at several depths using a probe that was specially designed for field use.

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®Lexan is a trademark of General Electric Co., Polymers Product Dept., Pittsfield, Massachusetts 01201.

Selected samples of sludge and underlying soil materials from the cores were subjected to total chemical analysis by X-ray fluorescence (XRF) analysis, and by inductively coupled argon plasma spectroscopy (ICAP) after dissolution. The mineralogy of selected sludge and soil subsamples was determined by X-ray diffraction (XRD) analysis. The sludge samples were also analyzed by scanning electron microscopy (SEM) and energy dispersive X-ray (EDS) analysis to identify entrapped fly ash particles and to confirm the presence of certain hypothesized minerals.

Pore waters were displaced from the sludge (by pressure) and soil cores (by immiscible displacement) under anoxic conditions, filtered (0.22- $\mu$ m and 0.0018- $\mu$ m filters), and stabilized with chemical preservation agents. Cations (Ba, Ca, K, Li, Mg, Na, and Sr) and metals (Al, Cd, Cr, Cu, Mn, Mo, Ni, Pb, Ti, V, and Zn) were determined on acidified (with Ultrex HCl) and nonacidified filtered samples by ICAP or, in a few cases, by graphite furnace atomic absorption. Silica and B were measured by ICAP. Anions ( $\text{SO}_4$ ,  $\text{S}_2\text{O}_3$ , SCN,  $\text{PO}_4$ ,  $\text{NO}_3$ , Br, Cl, I) and  $\text{SO}_3$  (with HCOH preservation) were analyzed by ion chromatography. The valence species of iron ( $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$ ) were quantified by ion chromatography. Ammonium ( $\text{NH}_4$ ) and F were analyzed by ion-selective electrode. Sulfide was determined by potentiometric titration with Pb. Organic carbon was measured with a carbon analyzer using persulfate-enhanced UV oxidation, and inorganic carbon was determined with acid sparging and IR detection.

## RESULTS AND DISCUSSION

Detailed investigations (4) were carried out to identify geochemical reactions controlling pore-water composition in the sludge and to evaluate the extent of solute migration into the underlying soils. These investigations included 1) field observations and measurements, 2) collecting and analyzing core samples from the waste materials and underlying soils, 3) performing interpretive experiments in the laboratory with fresh and aged FGD sludge and soil materials, and 4) applying a geochemical code for speciation and solubility calculations. The results of these investigations are briefly discussed under the headings Bulk Chemistry and Solid-Phase Characterization, Leachate Composition, and Solute Migration into Underlying Soil.

### BULK CHEMISTRY AND SOLID-PHASE CHARACTERIZATION

Because the sludge at this site was from a calcium carbonate scrubbing agent, it was hypothesized that the concentrations of all of the elements in the sludge other than Ca and C would originate from adsorption of gaseous flue gas species by  $\text{CaCO}_3$  or from the scrubbing of fly ash particles contained in the flue gas. Some variation in total chemical composition of samples from different locations in the sludge basin was therefore expected.

The range and mean for total elemental concentrations in the sludge are reported in Table 1, along with the range and mean concentrations reported for a variety of FGD wastes (5,6). Given the use of  $\text{CaCO}_3$  as the scrubbing agent at this plant, it is not surprising that the sludge is very high in Ca (27%) (Table 1). The other elements present in concentrations greater than 0.3% are Al, Ca, C, Fe, K, Mg, Na, P, S, Si, and Zn. The concentrations of many of these elements in this sludge are significantly different from those in other FGD wastes (Table 1). Most of the trace elements of interest, from an environmental standpoint, are present in small quantities. Arsenic, Cd, Cr, Cu, Ni, and V are present in concentrations less than 0.01%, and Ag, Hg, Mo, Sb, Se, and Sn are present in concentrations less than 0.001%.

Several samples from the sludge basin were analyzed by XRD to identify the crystalline components. The sludge samples were found to contain  $\text{CaCO}_3$  (calcite) and  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$  (hannebachite) with lesser amounts of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  (gypsum) and  $\text{SiO}_2$  (quartz). A few samples were found to contain  $\text{Ca}_4\text{Al}_2\text{CO}_3(\text{OH})_2 \cdot 5\text{H}_2\text{O}$ .

The samples were also examined by SEM. In general, photomicrographs (Figure 1) of the sludge show that it is composed of well-defined angular crystals with smaller amounts of spherical particles. The morphology of the spherical particles suggests that they are fly ash. Energy-dispersive X-ray spectroscopy was used to differentiate crystalline phases of  $\text{CaCO}_3$ ,  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$ , and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ . Calcite ( $\text{CaCO}_3$ ) was easily distinguished from the other crystalline phases because it contains Ca and C and does not contain S. The S/O ratios (1.75 for  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$  and 2.99 for  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) and the crystal habit were used to distinguish between  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$  and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ . Hannebachite ( $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$ ) is orthorhombic and frequently exhibits rectangular crystals (7) and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  is monoclinic and generally forms crystals that are non-rectangular parallelograms. Using these criteria, the grains with thin, flat, and rectangular shapes were identified as  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$ , and those with equant parallelogram shapes were identified as  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ . Analyses of a large number of the spherical particles showed them to be high in Fe and relatively high in Zn.

#### LEACHATE COMPOSITION

The sludge at this site contains large quantities of  $\text{CaCO}_3$  and  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$  and smaller amounts of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ . These three minerals have rapid precipitation/dissolution reaction kinetics. The similarity of observed ion activity products for  $(\text{Ca}^{2+})(\text{CO}_3^{2-})$ ,  $(\text{Ca}^{2+})(\text{SO}_3^{2-})$ , and  $(\text{Ca}^{2+})(\text{SO}_4^{2-})$ , calculated from the total aqueous concentrations in leachate samples by the geochemical model MINTEQA (8), with those calculated from independently available thermochemical data documented that  $\text{CaCO}_3$ ,  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$ , and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  control Ca and S concentrations in the leachates at this FGD sludge disposal site.

Field and laboratory methods were developed to accurately measure redox potentials. The redox potentials measured using these methods and those calculated from redox couples involving activities of individual S species were similar and found to represent chemically reducing conditions (about -300 mV at pH values between 8 and 9.4). Because of the very low redox potentials in the sludge, many redox-sensitive elements, such as As, Fe, and S (Table 2), were found or hypothesized to be in their reduced states. As a result of the reduced conditions and the presence of reduced S species, aqueous concentrations of many elements (e.g., Cd, Cr, Cu, Ni, Pb, Se, and Zn) were also maintained at very low levels and near their analytical detection limits (Table 2). Using a combination of direct (XRD and SEM) and indirect (comparison of ion activity products from samples and known solid phases) techniques, Rai et al. (4) have shown that the concentrations of these elements are controlled by solubility-controlling solids, inferred to be  $\text{CdS}$ ,  $\text{Cr}(\text{OH})_3$ ,  $\text{Cu}_2\text{S}/\text{Cu}_{1.65}\text{S}$ ,  $\text{NiS}$ ,  $\text{PbS}$ ,  $\text{FeSe}_2/\text{FeSe}$ , and  $\text{ZnS}$ . Given these solubility controls, the pore-water concentrations of these elements are not expected to change if reducing conditions continue to prevail.

Reducing conditions prevail in the sludge for a number of reasons. The reduced sludge has high moisture-retention characteristics and is underlain by a low-permeability soil, both of which inhibit drainage of the sludge water. As a result, the sludge is water-saturated through most of its depth in the basin. The water saturation, the abundance of  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$  and dissolved organic carbon, and the activity of sulfate-reducing bacteria have resulted in the chemically reduced sludge. The sulfate reducers, combined with other heterotrophic bacteria, are primarily responsible for the high sulfide conditions (Table 2) and the low redox potential in the sludge.



The high dissolved organic carbon (DOC) concentrations (Table 2) in the sludge pore waters promote the growth of heterotrophic bacteria that deplete dissolved oxygen from the pore water. Heterotrophic metabolism of the abundant DOC by the sulfate reducers continues under low-oxygen conditions, in which sulfate is used as the terminal electron acceptor (as oxygen would be under aerobic conditions). In accepting electrons, sulfate is reduced to sulfide and other reduced sulfur species. These reducing conditions prevail in the sludge because it is water-saturated and high in DOC. Oxygen diffusion into water-saturated sludge is minimal because of oxygen's slow molecular diffusivity in water-filled pores. In the sludge, oxygen is apparently replenished through diffusion more slowly than it is consumed by abiotic and biotic reductive processes.

It is surmised that the reducing conditions in the saturated sludge will prevail for several decades at least, because of expected high levels for microbiological populations of heterotrophs, sulfate reducers, and DOC. As a consequence, the leachate composition is expected to remain as it is now for several decades to come. If, however, the sludge drains, enhanced oxidation is anticipated because of the more rapid diffusivity of oxygen in gas-filled pore spaces. Oxygen, if present, will oxidize aqueous sulfide and solid-phase sulfides and retard the effects of sulfate-reducing bacteria. With oxidation, the amount of  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$  is expected to decrease with a corresponding increase in  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  and a slight increase in  $\text{CaCO}_3$ , corresponding to the increase in  $\text{CO}_2$  introduced from the air. The concentrations of Ca and  $\text{SO}_4$  in leachate are not expected to change with these changes in solid-phase composition, because the Ca and  $\text{SO}_4$  concentrations are currently in equilibrium with  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ , which is also the solid phase expected to form with oxidation. The pH values in the sludge with oxidation are still expected to be alkaline because of the presence of large quantities of  $\text{CaCO}_3$ . Under those oxidizing conditions, the sulfide solids of Cd, Cu, Fe, Ni, Pb, and Zn are not expected to be stable. The aqueous concentrations of many of these trace elements, however, are still expected to be controlled at low levels by their carbonate, hydroxycarbonate, hydroxide, or oxide solids.

To evaluate whether the assumed similarities between this study site and others that typify the electric power generation industry hold, the ion activity products of  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  observed at this site and those observed in sludge samples analyzed by Ainsworth and Rai (9) were compared (Figure 2). The ion activity products of  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  (Figure 2) at this site and in several other FGD waste samples are similar, and they are also similar to those in equilibrium with  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ . The results of X-ray diffraction and chemical analyses of solid samples from other power plants indicate the presence of  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$  and  $\text{CaCO}_3$ . Collectively, these results indicate that the sludge from this site is similar to other sludges with respect to at least the major elements and the major mineralogic phases.

The methods used to measure pH and Eh at this site were also applied to two others being sampled under the Leaching Chemistry Studies contract (EPRI RP2485-08). The pH and Eh values measured at these two other sites were similar to those found at this site and were very reducing. Microbiological analyses of these other two sites also showed the presence of large numbers of denitrifiers and sulfate-reducing bacteria. These in situ measurements, combined with pore-water analyses and microbiological enumerations, indicate that similar geochemical and biologic processes are operating at all sludge sites studied. The low redox potentials and the presence of sulfides in the samples ensure that metallic elements (such as Cd, Cu, Ni, Pb, and Zn) will be present in very low concentrations in pore waters or leachates. In support of this conclusion, it is noted that aqueous metal concentrations are extremely low at this field site, as well as in a larger set of FGD sludge samples analyzed (9) (Table 3).



In summary, based on limited data, it appears that the geochemical reactions that have been identified as controlling aqueous solute concentrations at this site are also important at other FGD sites.

#### SOLUTE MIGRATION INTO UNDERLYING SOIL

Scrubber sludge materials at the FGD waste site were disposed of directly on the surface of a fine-grained, clayey soil (Mollisol). The fact that FGD materials had overlain these soils for fifteen years provided a unique opportunity to assess the extent of solute migration from the sludge into the subsurface environment. This assessment was performed through chemical analysis of sludge, soil, and pore waters from continuous cores taken at the disposal site.

Assessment of solute migration required identification of the sludge-soil boundary underneath the pond. Visually, the interface was difficult to identify in the core samples because the highly reducing conditions at the base of the pile imparted a black color to both sludge and soil materials. Because the mineralogy of the sludge (calcium carbonate, calcium sulfate) differed from that of the underlying soils (aluminosilicates, quartz), the interface could be readily located using the total chemical analysis of Ca and Si by XRF (Figure 3). The soil-sludge interface is identified by the inflection in the total Ca and Si concentrations. This technique was effective at two coring locations in the basin where the sludge depth differed (i.e., 5 m and 2.5 m). This analytical approach to quantify the sludge-soil boundary is expected to be equally effective at other FGD sites except for those underlain by limestone or similar calcareous materials.

The total concentrations of As, B, Cd, Pb, Se, Sr, and Zn measured by XRF were higher in the sludge solids than in the underlying soils. Zinc was especially elevated. With the exception of B, however, the total chemical analysis showed that these elements exhibited negligible mobility across the sludge-soil boundary (Figure 4). Geochemical processes within the sludge, notably precipitation/dissolution, were found to be especially important in immobilizing the metallic elements. For example, experimental evidence suggested that metal sulfides in the sludge maintain aqueous concentrations of Zn, Cd, and Pb at the analytical detection limit. In contrast, the total chemical analyses of B demonstrated no inflection at the soil-sludge boundary (Figure 4), suggesting it had migrated from the sludge.

Analyses of pore waters from sludge and soil corroborate the results of the total chemical analysis. Metallic elements including Zn, Cd, and Pb were below the detection limit in all pore-water samples from the soils, consistent with their hypothesized immobilization in the sludge by formation of sulfide solids. Cationic solutes, including  $\text{Sr}^{2+}$ ,  $\text{K}^+$ , and others that are elevated in sludge pore waters relative to nearby soils, were found to be rapidly removed (attenuated) from the infiltrating sludge leachate by ion exchange to clay minerals in the fine-grained soil. Boron was observed to be the most mobile solute from the sludge (Figure 5), moving to a depth of nearly 1 m below the sludge-soil interface. The steep concentration profile of B was interpreted as being a result of the limited infiltration volume of sludge leachate, combined with chemical attenuation through adsorption by the clay minerals.

Boron was found to be the best tracer of sludge-water infiltration, and a retardation factor analysis combined with moisture content measurements of the underlying soil placed the maximum infiltration depth of leachate at 2-3 m. Minimal solute migration was therefore observed because of the limited permeability of the clayey soil and its high attenuation capacity for cationic solutes released from sludge.

## SUMMARY AND CONCLUSIONS

A field investigation of a decommissioned FGD sludge pond was performed to assess both the long-term geochemical processes within the sludge and solute migration into the underlying subsurface environment. The total concentrations of most elements in the sludge fell within the range observed in a previous survey of FGD sludge wastes (9). Certain metallic elements (such as Pb and Zn), however, were higher in total concentration in the sludge at this site because significant quantities of fly ash were incorporated into this waste during the flue gas scrubbing process. Pore-water concentrations were found to be homogeneous in the sludge and were highly reducing as a result of reduced S species and of microbiological activity. Precipitation/dissolution reactions with  $\text{CaCO}_3(\text{s})$ ,  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}(\text{s})$ , and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}(\text{s})$  appeared to control the macroion concentrations in the sludge pore waters. These same types of reactions with sulfides limited dissolved concentrations of Cd, Cu, Ni, Pb, and Zn to levels that were below detection limits.

Leachate migration from the sludge basin was minimal because the underlying soils were clayey and low in permeability and exhibited significant attenuation capacity for many of the elements in the sludge pore water. Boron was weakly adsorbed by the underlying subsoils and, for this reason, was found to be the best tracer of sludge-water infiltration. At the time of this study, B had migrated approximately 1 m into the underlying subsoil. This observation, combined with laboratory adsorption measurements, allowed us to estimate that sludge water had infiltrated approximately 3 m into the subsoil over the pond's 15-year lifetime.

## NOTICE

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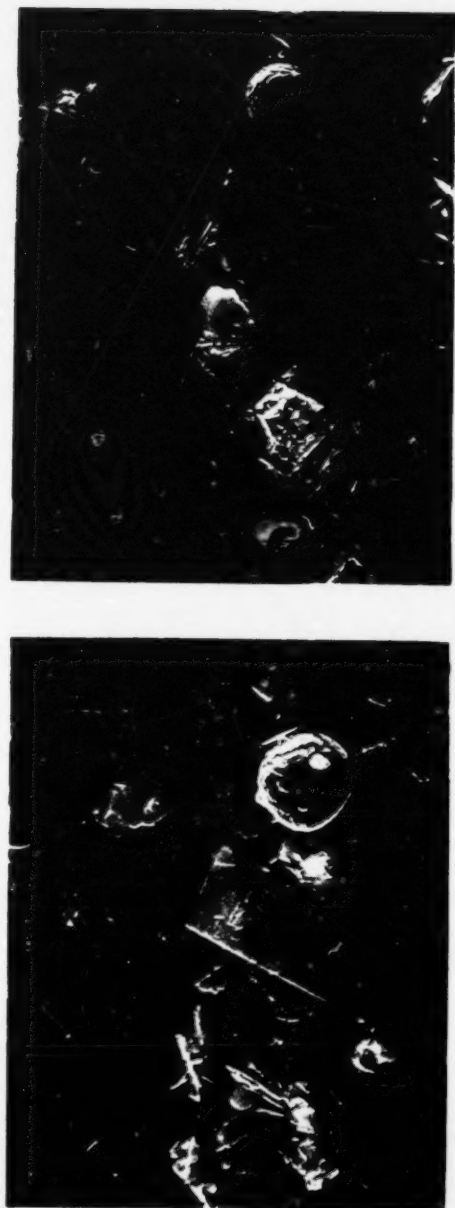


Figure 1. Photomicrographs of sludge samples. The spherical particles are probably from entrapped fly ash, and they are in most cases rich in Fe and relatively rich in Zn (about 50% Fe and up to as much as 20% Zn). Predominant crystalline minerals are  $\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$  (hannebachite) and  $\text{CaCO}_3$  (calcite). Typical crystals of hannebachite are thin and rectangular (see top left photo); typical calcite crystals are blocky with surface overgrowths (see top right photo). A crystal with parallellogram shape (bottom photo) may be  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  (gypsum).



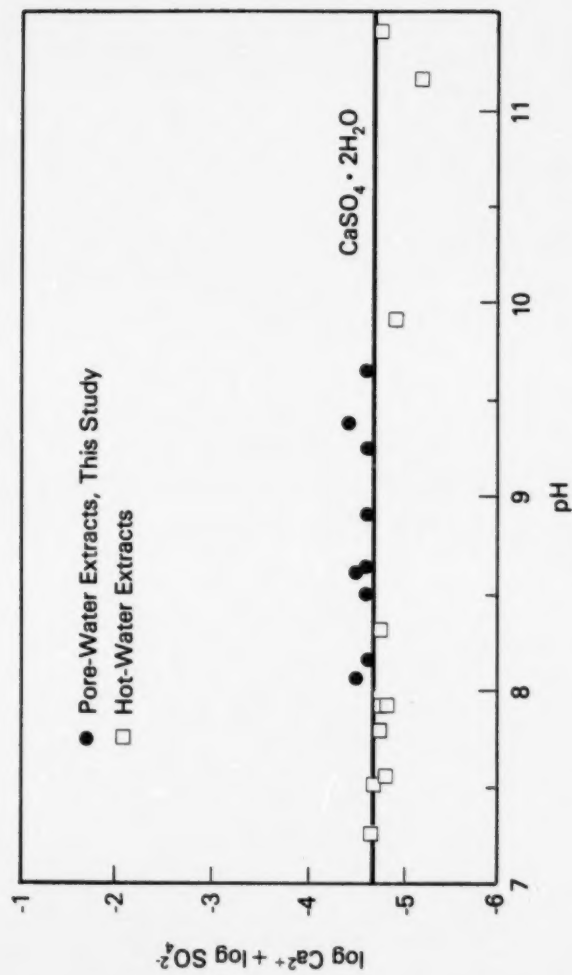


Figure 2. Comparison of ion activity products of  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  in pore-water extracts (this study, shown as solid circles) and hot-water extracts from sludge samples from different geographic locations (Ref. 9, shown as open squares). Solid line represents ion activity product of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ .

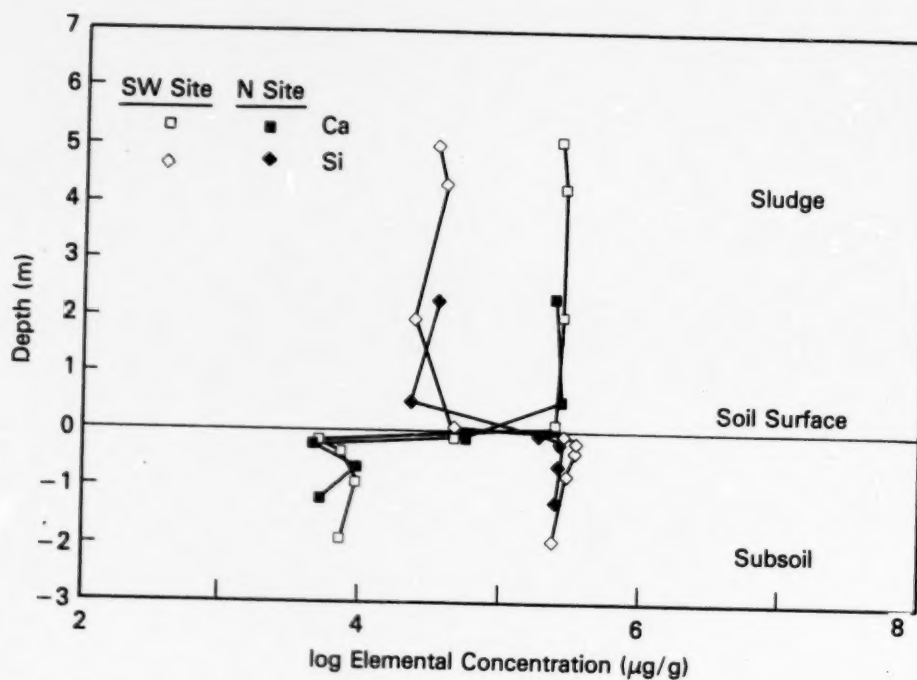


Figure 3. Total concentrations of Ca and Si in sludge and underlying soil at the SW and N coring locations.

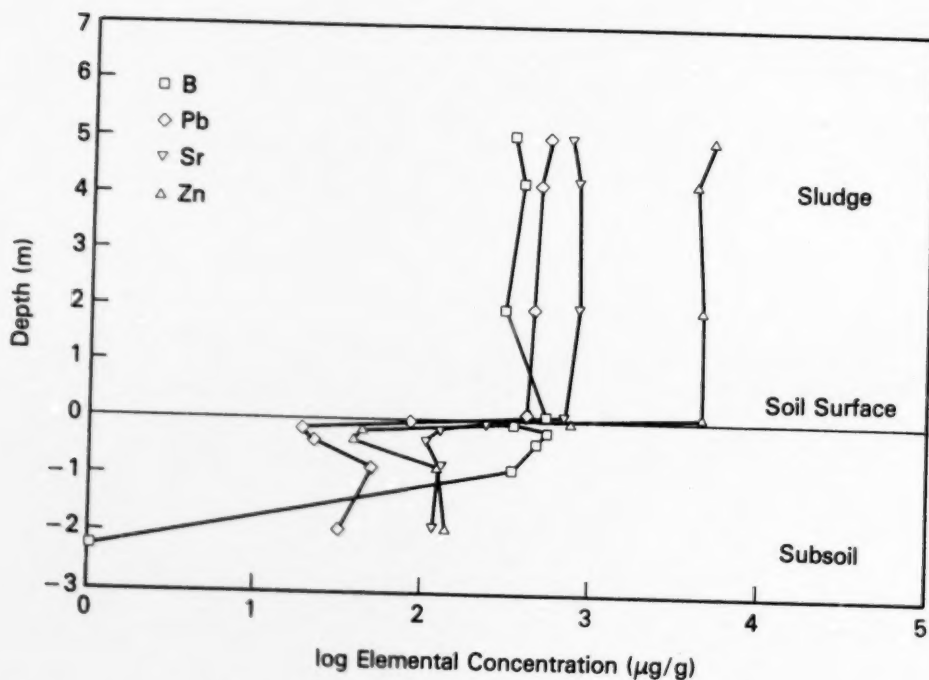


Figure 4. Total concentrations of B, Pb, Sr, and Zn in sludge and underlying soil at the SW coring location.

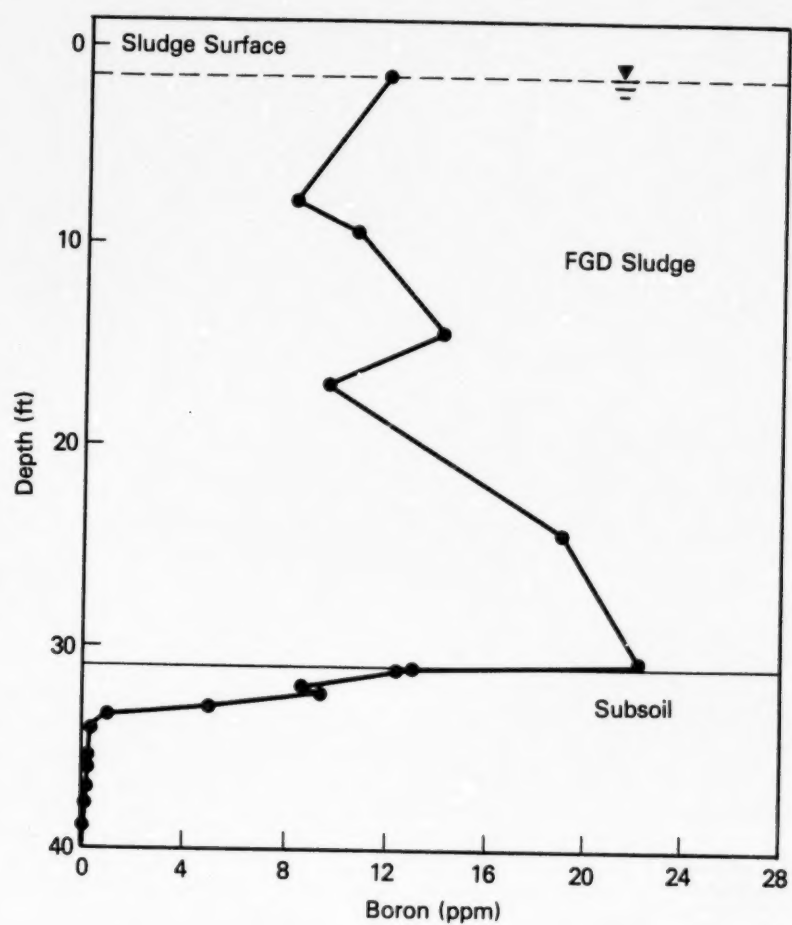


Figure 5. Concentration of B in pore waters as a function of depth in FGD sludge and underlying soil.

TABLE 1. RANGE AND AVERAGE TOTAL CONCENTRATION OF ELEMENTS IN SELECTED SAMPLES OF FLUE GAS DESULFURIZATION WASTES

| Element   | Concentration $\mu\text{g/g}$ Solid (Wet Process Wastes) |                 |         | Concentration $\mu\text{g/g}$ Solid (Dry Process Wastes) |                     |                 |
|-----------|----------------------------------------------------------|-----------------|---------|----------------------------------------------------------|---------------------|-----------------|
|           | This Study*                                              |                 | Mean    | Furnace                                                  |                     | Mean            |
|           | Mean                                                     | Range           |         | Sorbent Injection Wastes†                                | Spray Dryer Wastes‡ |                 |
|           |                                                          |                 |         | Mean                                                     | Range               | Range           |
| Al        | 14,229                                                   | 11,700-17,200   | 30,300  | 52,080                                                   | 43,500-79,000       | 33,300-66,700   |
| Ag        | 7.8                                                      | <6.7-10.3       | --      | 2.6                                                      | 1.8-3.5             | 1.6-5.8         |
| As        | 71.5                                                     | 59.1-88.9       | 25.2    | 37.4                                                     | 15-62               | 4.2-48          |
| B#        | 120.6                                                    | 80##-467.5      | --      | 52.8                                                     | <10-82              | <10-230         |
| Ba        | 96                                                       | 50-150          | 370     | 290                                                      | 220-450             | 190-6,060       |
| C (inorg) | 42,723                                                   | 23,420-60,250   | --      | --                                                       | --                  | --              |
| Ca        | 271,714                                                  | 258,000-300,000 | 274,000 | 215,580                                                  | 120,000-285,700     | 126,200-223,000 |
| Cd        | 63.3                                                     | 44.6-81.9       | <5      | 0.66                                                     | <0.1-1.8            | --              |
| Cl        | 417                                                      | <340-480        | 2,514   | --                                                       | --                  | --              |
| Cr        | 63                                                       | <41-96          | 72.7    | 88.8                                                     | 77-100              | 36-210          |
| Cu        | 55.4                                                     | 47.1-59.3       | 31.8    | 41.4                                                     | 22-77               | 20-160          |
| Fe        | 27,793                                                   | 19,150-41,000   | 64,000  | 50,840                                                   | 41,700-59,000       | 10,900-53,700   |
| Hg        | 10.4                                                     | <9.5-11         | --      | 0.59                                                     | 0.45-0.93           | 0.1-0.4         |
| K         | 4,094                                                    | 3,480-4,620     | 1,900   | 4,064                                                    | 4,500-12,300        | 1,760-7,020     |
| Mg#       | 3,867.8                                                  | 1,975.5-4,619   | 7,200   | 3,676                                                    | 3,760-15,000        | 3,880-27,700    |
| Mn        | 373                                                      | 304-483         | 138     | 166                                                      | 140-240             | 55-680          |
| Mo        | 8.6                                                      | 5.6-13.4        | 12.2    | 5.9                                                      | 3.3-9.8             | 2.7-514         |
| Na        | 7,598                                                    | 4,860-11,080    | 3,817   | 4,030                                                    | 1,790-6,560         | 6,540-34,000    |
| Ni        | 77.2                                                     | 68-98           | 40.4    | 75.4                                                     | 44-110              | 17-110          |
| P         | 4,086                                                    | <2,100-6,200    | 4,271   | --                                                       | --                  | --              |
| Pb        | 452                                                      | 396-527         | 29.2    | 13.2                                                     | 2.4-42              | <0.3-14         |
| S         | 132,329                                                  | 77,600-189,800  | 152,000 | --                                                       | --                  | --              |
| Sb        | 10.9                                                     | <9.8-15.5       | 15.5    | 19.8                                                     | <12-24              | <5.6-29         |
| Se        | 3.8                                                      | 2.8-5.3         | 35.8    | 4.4                                                      | <0.4-17             | <0.4-7.3        |
| Si        | 32,486                                                   | 20,900-39,700   | 66,000  | 90,380                                                   | 73,500-130,000      | 74,500-120,000  |
| Sn        | 8.6                                                      | <8.3-48.9       | 29.9    | 79.2                                                     | <24-130             | 24-349          |
| Sr        | 753                                                      | 694-801         | 608     | 432                                                      | 180-790             | 184-3,040       |
| V         | 71                                                       | 58-97           | 156     | 152                                                      | 100-190             | 72-180          |
| Zn        | 4,240                                                    | 3,820-5,070     | 99.7    | 57.4                                                     | 26-76               | <6-79           |
| pH        | --                                                       | --              | --      | 12                                                       | 11.8-12.5           | 7.7-12.0        |

\*Based on 7 sludge samples from different depths and locations in the sludge basin.

†Based on 10 sludges from 8 power plants.

‡Based on 5 samples from pilot-scale units.

#Based on 8 samples.

#Concentrations were determined by matrix dissolution with NaOH or KOH followed by solution analysis using inductively coupled plasma-atomic emission spectroscopy (ICP).

##80 = below detection level.



TABLE 2. RANGE AND AVERAGE AQUEOUS CONCENTRATION OF ELEMENTS  
IN FGD SLUDGE PORE WATERS

| Species                       | Concentration (mg/L)<br>Average (Range)* |
|-------------------------------|------------------------------------------|
| Al                            | 0.38 (<0.3 - 1.03)                       |
| As(III)                       | 1.21 (0.02 - 2.66)                       |
| As(V)                         | 0.26 (0.03 - 0.45)                       |
| B                             | 15.4 (9.41 - 22.4)                       |
| Ba                            | 0.07 (0.05 - 0.10)                       |
| Br <sup>-</sup>               | 3.11 (1.2 - 6.1)                         |
| C (inorg.)                    | 1.45 (0.48 - 4.13)                       |
| C (org.)                      | 145.5 (73.7 - 217.3)                     |
| Ca                            | 743 (679 - 788)                          |
| Cd                            | 0.0011 (<0.0007 - 0.002)                 |
| Cl <sup>-</sup>               | 466 (194 - 932)                          |
| Cr                            | 0.0013 (<0.001 - 0.003)                  |
| Cu                            | 0.011 (<0.010 - 0.015)                   |
| F <sup>-</sup>                | 6.87 (5.66 - 7.72)                       |
| Fe <sup>2+</sup>              | 0.073 (0.005 - 0.21)                     |
| Fe <sup>3+</sup>              | 0.050 (0.010 - 0.140)                    |
| I <sup>-</sup>                | 11.52 (2.1 - 29.4)                       |
| K                             | 162 (47 - 431)                           |
| Li                            | 0.70 (0.50 - 0.88)                       |
| Mg                            | 7.53 (<0.6 - 28.20)                      |
| Mn                            | 0.0739 (0.0034 - 0.33)                   |
| Mo                            | 3.47 (0.48 - 8.58)                       |
| Na                            | 136 (75 - 244)                           |
| Ni                            | 0.017 (<0.01 - 0.04)                     |
| NH <sub>4</sub> <sup>+</sup>  | 4.34 (0.62 - 10.64)                      |
| NO <sub>2</sub> <sup>-</sup>  | 7.285 (0.004 - 49.7)                     |
| NO <sub>3</sub> <sup>-</sup>  | 7.73 (0.19 - 48.1)                       |
| Pb                            | 0.012 (<0.01 - 0.028)                    |
| PO <sub>4</sub> <sup>3-</sup> | 0.36 (0.14 - 0.5)                        |
| Se                            | <0.01                                    |
| Si                            | 6.77 (2.4 - 11.6)                        |
| S <sub>2</sub> O <sub>3</sub> | 95.2 (3 - 215)                           |
| S <sub>2</sub> <sup>-</sup>   | 3.1 (<0.3 - 14.2)                        |
| SO <sub>3</sub> <sup>2-</sup> | 19.3 (16.1 - 21.4)                       |
| SO <sub>4</sub> <sup>2-</sup> | 1484 (1415 - 1598)                       |
| Sr                            | 7.727 (6.82 - 9.2)                       |
| V                             | 0.044 (0.025 - 0.087)                    |
| Zn                            | <0.02                                    |

\*Based on nine samples. The lower-range values preceded by <, and the values given for Se and Zn, are the analytical detection limits for those species. The average values for Al, Cd, Cr, Cu, Ni, and V are near their detection limits. The values for all of the other elements are well above the detection limits.

TABLE 3. AVERAGE AQUEOUS CONCENTRATIONS OF SELECTED TRACE ELEMENTS IN DIFFERENT SLUDGES

| Element | Concentration (mg/L) in Different Sludge Samples |                |        |       |        |       |        |        |        |        |    |
|---------|--------------------------------------------------|----------------|--------|-------|--------|-------|--------|--------|--------|--------|----|
|         | This Study+                                      | Other Samples* |        |       |        |       |        |        |        |        |    |
|         |                                                  | 1              | 2      | 3     | 4      | 5     | 6      | 7      | 8      | 9      | 10 |
| Cd      | <0.001                                           | 0.005          | <0.002 | 0.005 | <0.002 | 0.006 | <0.002 | <0.002 | <0.002 | <0.002 |    |
| Cu      | <0.01                                            | 0.007          | <0.004 | 0.006 | 0.008  | 0.007 | <0.004 | 0.005  | 0.02   | 0.01   |    |
| Ni      | <0.01                                            | 0.03           | <0.01  | <0.01 | 0.02   | <0.01 | <0.01  | <0.01  | <0.01  | <0.01  |    |
| Pb      | <0.01                                            | 0.08           | <0.06  | <0.06 | <0.06  | <0.06 | <0.06  | <0.06  | <0.06  | <0.06  |    |
| Zn      | <0.02                                            | 0.04           | <0.01  | <0.01 | <0.01  | <0.01 | <0.01  | <0.01  | <0.01  | <0.01  |    |

\*Samples from different geographic locations, produced at different coal-fired power plants (9).  
 +Average of 7 pore-water extracts.

## SPRAY DRY ABSORPTION-OXIDATION OF SO<sub>2</sub>/NO<sub>x</sub>

By: Ann-Mari Strömberg and Hans T. Karlsson  
Department of Chemical Engineering II  
University of Lund  
S-221 00 Lund, Sweden

### ABSTRACT

An experimental investigation has been conducted in order to elucidate the feasibility of simultaneous removal of SO<sub>2</sub> and NO<sub>x</sub> by spray dry absorption. To this end, an extensive screening program was carried out. The objective was to identify additives that would enable NO<sub>x</sub> removal by minor modifications of the process chemistry of conventional lime based spray dry absorption. The most promising candidates were tested in a bench scale system consisting of a spray dryer and a fabric filter. A modified absorption-oxidation system with sodium chlorite was found to remove NO<sub>x</sub> most efficiently. The additive was shown to work for both lime and limestone, yielding up to 75 percent NO<sub>x</sub> removal. The additive is expensive. However, the test program has demonstrated the technical feasibility of simultaneous removal of NO<sub>x</sub> and SO<sub>2</sub> in spray dry absorption. The dry end product consists of calcium sulfate, calcium nitrate, sodium chloride and unreacted alkali. The bench scale tests were verified in pilot plant equipment.

### INTRODUCTION

#### BACKGROUND

Spray dry scrubbing is a well established method for the removal of SO<sub>2</sub>. Currently, more than 50 such systems are in operation around the world. In most of the systems a lime slurry is sprayed into a spray dryer, and the dry product is captured in a downstream fabric filter. Concurrently, the flue gas temperature decreases due to water evaporation. The SO<sub>2</sub> is removed both in the spray dryer and in the fabric filter system.

Since there are increasing requirements to reduce the NO<sub>x</sub> emissions from utility boilers, there is also an increasing demand for the removal of both SO<sub>2</sub> and NO<sub>x</sub> from flue gases. This can be achieved either by utilizing two separate processes, or simultaneously in one process. The conventional lime based spray dry scrubbing process for removal of SO<sub>2</sub> hardly removes any NO<sub>x</sub> at all. It would obviously be advantageous if SO<sub>2</sub> and NO<sub>x</sub> could be removed simultaneously in a spray drying process. In fact, there are also two such simultaneous spray drying techniques under development; in one case, sodium hydroxide is added to the lime slurry to enhance the NO<sub>x</sub> removal [1]; the other concept is based on zinc oxide as an absorbent for both SO<sub>2</sub> and NO<sub>x</sub> [2].

The leading technique used today to control SO<sub>2</sub> and NO<sub>x</sub>, is a combination of flue gas desulfurization (FGD) and selective catalytical reduction (SCR) of NO<sub>x</sub> with ammonia. There are also several wet and dry processes available for simultaneous removal of SO<sub>2</sub> and NO<sub>x</sub>; 90 percent removal efficiency can be achieved for both SO<sub>2</sub> and NO<sub>x</sub> [3]. Most of these processes are still under development, but there are some processes in commercial operation. The Walter process removes SO<sub>2</sub> with ammonia in a scrubber, at the same time NO<sub>x</sub> is removed by oxidation of NO to NO<sub>2</sub>, which in turn

is absorbed by the ammonia solution. Another system has been developed by Bergbau-Forschung. In this process,  $\text{SO}_2$  is adsorbed on activated coke, and  $\text{NO}_x$  is simultaneously reduced with ammonia.

In the wet simultaneous processes under development, alkali is often used together with oxidizers such as  $\text{ClO}_2$ ,  $\text{KMnO}_4$ ,  $\text{NaClO}_2$  or  $\text{H}_2\text{O}_2$ . The most developed wet process from a commercial point of view uses a scrubbing liquor containing  $\text{Fe/EDTA}$ . The dry processes under development are primarily based on a variety of principles, including the use of catalysts, ammonia or electron beams.

The study presented in this paper was undertaken to extensively investigate the removal efficiency of  $\text{NO}_x$  and  $\text{SO}_2$  in a lime based spray dry scrubbing process by applying a modified process chemistry. The modification consisted of an alteration of the chemistry which was accomplished by using an additive. The additive selection was based on the results from a screening program of a number of candidate additives.

Experiments were performed in a spray dry scrubbing system of bench-scale size, consisting of a spray dryer followed by a fabric filter. Slurries were prepared from lime or limestone as basic absorbents which was supplemented with the additive. Further tests were done in pilot plant equipment to confirm the small-scale tests.

### SCREENING TESTS

A comprehensive screening program has been carried out in order to identify suitable chemicals which have the potential to enable the removal of  $\text{NO}_x$  in a spray dry scrubber. The desired goal was to find a substance that would act as a promotor in small amounts when added to a conventional lime slurry. Such a substance used as an additive should of course be inexpensive and pose no additional problem in a spray dry scrubbing process. This latter problem includes all aspects of the dry end product. However, it was also decided that more sophisticated and expensive substances were of interest for testing in the screening program.

Candidate additives were selected based on previous information on the chemistry of  $\text{NO}_x$ , which includes basic reactions, as well as dry and wet systems previously investigated in the literature and at our department. Due to the nature of a spray dry process, reactions under both wet and dry conditions are obviously of interest. It is known that  $\text{NO}_x$  can undergo a large number of reactions, which has been extensively reported in the literature.

Table 1 displays some 40 additives that were selected for further tests. The additives were mixed with lime on a five mole-percent basis. The mixtures were then screened in the sand bed reactor for their ability to remove  $\text{NO}_x$  simultaneously with  $\text{SO}_2$ . Each additive was in general tested five times under standard conditions. The sand bed reactor was developed in order to obtain a simple device for the investigation of reactions between gas and solids under bagfilter conditions; such conditions pose large experimental problems due to the small particles subjected to investigation [4]. The sand bed reactor has been extensively tested ([5], [6]), and it has been accepted by several research groups as a general tool to investigate gas solids reactions under bagfilter conditions [2], [7], [8], [9].

Table 1 displays the observed capabilities of the additives for the removal of  $\text{SO}_2$ ,  $\text{NO}$  and  $\text{NO}_2$ , respectively, under typical flue gas conditions. Of the least sophisticated additives, a mixture of urea and sodium hydroxide was found to be the best candidate. This additive was then further tested in roughly 50 additional experiments in the sand bed reactor in order to identify the optimal molar ratios urea/ $\text{NaOH/Ca(OH)}_2$ . Further details on this have been presented elsewhere [10]. However, the most promising additive show only very small effects when tested in a 0.5  $\text{MW}_{\text{th}}$  spray dry scrubber. Not more than 25 percent removal efficiency of  $\text{NO}_x$  was obtained. Virtually all of the removal was obtained in the fabric filter.

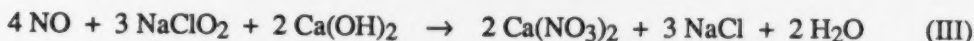
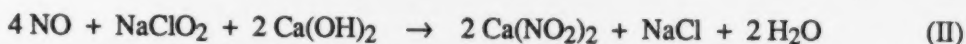
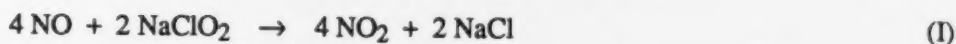
Further tests were then undertaken with more expensive additives. The most reactive absorbents for  $\text{NO}_x$  under wet conditions are known to be  $\text{NaClO}_2$ ,  $\text{KMnO}_4$  and  $\text{Fe}^{2+}/\text{EDTA}$ , [11]. The kinetics for these systems have been extensively studied for more than a decade [12], [13], [14]. Furthermore, two of the systems were recently suggested for use in spray dry scrubbing [15]. The screening tests revealed that sodium chlorite ( $\text{NaClO}_2$ ) should be of greatest interest as an additive. The further work described in this paper was based on this additive.



## THEORETICAL CONSIDERATIONS

### CHEMICAL REACTIONS

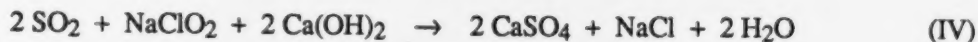
Several kinetic studies have been carried out over the past decade to investigate the wet absorption of NO and NO<sub>2</sub> in sodium chlorite solutions. In all cases, very simple overall chemical reactions have been assumed. However, the reaction pattern is quite complex and clearly not completely outlined. Starting with NO, which is considered to be the major NO<sub>x</sub> component in combustion flue gases, oxidation to NO<sub>2</sub>, NO<sub>2</sub><sup>-</sup> and NO<sub>3</sub><sup>-</sup> may occur. Furthermore, these latter products may be intermediates subjected to further oxidation. Overall reactions to consider in a lime/chlorite system are then:



It is obvious from these reactions that one mole of chlorite has the ability to oxidize from 4/3 up to 4 moles of NO, depending on the actual reaction pattern. Obviously, if NO is oxidized to NO<sub>2</sub>, no NO<sub>x</sub> is captured in the system unless NO<sub>2</sub> is further oxidized.

The absorption of NO consumes alkali, corresponding to 0.5 moles of calcium for each mole of NO absorbed. Furthermore, the concentration of hydroxide ion plays an important role in determining which reaction pattern will dominate. This is because hydroxide affects the reaction rate for the reactions in different ways.

If SO<sub>2</sub> is present in the system, the reaction pattern is complicated. However, one major overall reaction to be considered is the oxidation of sulfite to sulfate. So, the following reaction should be expected to consume chlorite to some extent:



Thus, each mole of chlorite can oxidize two moles of sulfite.

### REACTION KINETICS

It is generally known that NO only reacts at a high rate with a limited number of species. For most NO reactions the rate is slow. In a spray dry scrubber both the dry reaction in the fabric filter and the wet reaction in the spray dryer have to be considered. The screening tests used as a basis for the selection of an additive were performed in the sand bed reactor, thus revealing only the potential for the dry reaction. However, there are two reasons for performing tests in a sand bed reactor. First, the experiments are easy to perform, which enables a large number of candidates to be tested. Second, it is generally known, as previously mentioned, that fast wet NO reactions are scarce.

It is clear that bagfilter conditions cannot be exactly simulated in the sand bed reactor. Most important is that a bagfilter has a spray dryer ahead, so the reactants are always partly reacted when entering the fabric filter. Therefore, the screening is intended only to give a ranking of the candidate additives. However, the conditions in the sand bed reactor were set to reflect the conditions of a full scale unit, in which case parameters such as the temperature and the relative humidity have to be considered. Furthermore, by utilizing a correct ratio of the mass of lime to the gas flow rate, the actual removal data should give some indications of anticipated performance in a fabric filter. A typical value of the ratio mentioned is 20 g lime per Nm<sup>3</sup>/h. The results for pure lime indicated less than 20 percent NO<sub>x</sub> captured as an overall value for 30 minutes. In the presence of chlorite, 100 percent NO<sub>x</sub> removal was seen [10]. This gives at least a fair indication.

At first glance, analyzing the parameters for the wet conditions is very complex, because of the greater number of processes occurring simultaneously during the drying of fine spray. However, a very simple and exact analysis can be done by applying an analogy between NO and SO<sub>2</sub>. Such an analysis can be used directly to outline the prospects for various absorbents or additives by simply knowing the value of a single rate constant. By applying basic theory, the local value of the mass transfer rate of NO can be derived for the gas phase and the liquid phase, respectively, according to the following identity:

$$N_A = \frac{D_{Ag}}{R} (C_{Ag} - C_{Ai}) = (E_A - 1) \frac{D_{Al} R T}{R H_A} C_{Ai} \quad (1)$$

It is known from previous studies, [16] that substantial mass transfer resistance must prevail in the gas phase in order to enable any substantial amounts of removal of a gaseous species in a spray dryer. The fraction of gas phase resistance can be derived from the previous equation:

$$F_A = 1 - \frac{C_{Ai}}{C_{Ag}} = 1 - \left[ 1 + \frac{D_{Al} R T}{D_{Ag} H_A} (E_A - 1) \right]^{-1} \quad (2)$$

In order to apply an analogy with SO<sub>2</sub>, we use the expression for mass transfer in the gas phase for this species:

$$N_B = \frac{D_{Bg}}{R} (C_{Bg} - C_{Bi}) \quad (3)$$

For substantial removal of NO it is alledged that the fraction removal rate of NO should be as high as the fraction removal rate of SO<sub>2</sub>. Hence:

$$\frac{N_A}{C_{Ag}} = \frac{N_B}{C_{Bg}} \quad (4)$$

Under certain conditions, a lime based system approaches complete gas phase control for SO<sub>2</sub>, eg. when using lime in excess [16]. This means that  $C_{Bi}$  approaches zero. If the mass transfer of NO is studied under such conditions, we obtain the following simply by combining the equations above:

$$F_A = \frac{D_{Bg}}{D_{Ag}} \quad (5)$$

This parameter is virtually a constant, and the diffusion coefficients can be estimated from standard text books, yielding  $F_A = 0.62$ . This value can now be used to estimate the required enhancement factor for NO. As can be seen from Equation (2), only a number of constants need to be estimated for this purpose. The constants should be calculated at the adiabatic saturation temperature (55 °C), which is the temperature the droplets attain quite quickly in a spray dryer. Data for the constants inserted in Equation (2) lead to an enhancement factor of  $E_A = 3 \cdot 10^5$ .

This estimated value can now be compared to values measured in small scale laboratory tests. Studying the most reactive species known, ie. NaClO<sub>2</sub> and KMnO<sub>4</sub>, enhancement factors of the order of  $E_A = 10^4$  have been obtained experimentally [13], [17]. Even though the experiments were performed at quite a low temperature the data indicate the magnitude of the problem of absorbing NO in a spray dry scrubber;  $E_A$  increases with the temperature. If it is assumed that the enhancement factor is an order of magnitude lower than the calculated value, as shown by the experiments, Equation (2) indicates that the fraction removal rate of NO is 20 percent of the fraction removal rate of SO<sub>2</sub>.

It is not completely correct to compare the enhancement factors obtained from two different types of equipment. The analysis can then be carried out a little bit further to circumvent this problem. If excess of sodium chlorite prevails, the reaction with NO is of pseudofirst order. The enhancement factor for a pseudofirst order reaction occurring in a droplet can simply be written as:

$$E_A = \frac{\gamma}{\tanh \gamma}; \quad \gamma = R \sqrt{\frac{k}{D_{AI}}} \quad (6)$$

Using this equation and having a typical sauter drop diameter of 25 microns [16] leads to an estimated rate constant of the order of  $10^{12} \text{ s}^{-1}$ . This value is several orders of magnitude higher than, for instance, the rate constant for hydrolysis of  $\text{SO}_2$ , which in general is considered to be an instantaneous reaction.

## EXPERIMENTAL

### SPRAY DRY ABSORBERS

A schematic presentation of the bench scale spray dry scrubber used in this study is shown in Figure 1 and a data sheet is given in Table 2. More extensive descriptions are given elsewhere [19], [20]. Simulated flue gas is obtained by mixing gases from cylinders through pressurized, calibrated rotameters. The inert components of the gas are bubbled through a tempered water bath, the humidifier, to reach the desired moisture content of the flue gas. The gas mixture is then led through a preheater packed with Raschig rings to heat the gas to the desired inlet temperature of the spray dryer.

The spray dryer and the baghouse are provided with water jackets to keep the system under adiabatic conditions. The flue gas enters the top of the dryer where the slurry is also injected by an ultrasonic nozzle. The water evaporates when the atomized slurry comes in contact with the hot flue gas and the dry product is separated from the flue gas in the baghouse. The baghouse contains five bagfilters, each provided with shut-off valve. The filters are cleaned with jet pulses. Gas analyses are done with a Binos UV-instrument ( $\text{SO}_2$ ) and a Beckman chemiluminescence instrument ( $\text{NO}_x$ ).

Studies were also carried out in a pilot plant, treating flue gas with a flow rate of  $800 \text{ Nm}^3/\text{h}$ , corresponding to about 0.5 MW heating rate. A schematic drawing of the spray dry scrubbing pilot plant is shown in Figure 2 and a detailed data sheet is given in Table 2. The pilot plant is operated on a single pass basis with no recycle of the spent absorbent.

The pilot plant consists of a spray dryer equipped with a two-fluid nozzle for slurry atomization and a jet pulse fabric filter. Flue gas is generated by burning low sulfur oil to heat air to the desired spray dryer inlet temperature. Pure  $\text{SO}_2$  is added from cylinders using a calibrated rotameter. Similarly,  $\text{NO}_x$  is added by taking pure  $\text{NO}_2$  from a cylinder and running the gas through a thermally heated reactor, the converter, where most of the  $\text{NO}_2$  is reduced to NO. The adiabatic saturation temperature is held constant by adding live steam to the gas stream. The flue gas is introduced at the top of the spray dryer.

The slurry is prepared in a stirred storage tank. Slurry and water are fed to the nozzle via a computerized weight sensor system. The slurry feed rate is set at the desired stoichiometric ratio while the water feed rate is set at the desired spray dryer outlet temperature. A specially designed two-fluid nozzle is used for atomization of the slurry with compressed air. The nozzle gives a major droplet fraction below 100 microns and a typical sauter mean diameter of about 20 to 30 microns [16].

The particulate removal device is a Flakt Optipuls baghouse, containing twenty bagfilters with a total area of  $11.2 \text{ m}^2$ . The bagfilters are cleaned with high pressure jet pulses. The flue gas is discharged through a stack by an induced draft fan located downstream from the baghouse. Gas samples are continuously withdrawn from the flue gas between the spray dryer and the baghouse and after the baghouse for analysis of  $\text{SO}_2$ , using two URAS 2G IR-instruments, and for analysis of  $\text{NO}_x$ , using a Beckman chemiluminescence instrument. The gas sample between the spray dryer and the fabric filter is passed through a particle filter which is heated to  $200^\circ\text{C}$ . Reactions between  $\text{SO}_2$  and the particles are negligible at this temperature because of the low relative humidity [16].

### EXPERIMENTAL PROCEDURE

The study was primarily focused on the use of slurries prepared from slaked lime, because this was the basic absorbent used in the screening tests. A commercial dry slaked lime was utilized in the test



runs. However, experiments were also made with a commercial limestone with a particle size of 100 percent less than 200 microns. The properties of the two sorbents are displayed in Table 3. Furthermore, a commercial type of sodium chlorite with 80 percent by weight of  $\text{NaClO}_2$  was utilized as an additive.

The major part of the experiments were performed in the bench scale equipment using the experimental conditions shown in Table 2. During all these experiments, the fabric filter was operated at a very low pressure drop in order to be able to collect a large number of data points. The removal of  $\text{SO}_2$  and  $\text{NO}_x$  were recorded under steady state conditions for both the spray dryer and the fabric filter. The flue gas entering the spray dryer contained only NO as an  $\text{NO}_x$  species, while obviously both NO and  $\text{NO}_2$  were recorded at the exit of the spray dryer and the fabric filter.

The impact of the amount of added sodium chlorite was first investigated under baseline conditions. The relative humidity of the flue gas exiting the fabric filter was set at 62 or 40 percent. This corresponds to 9 °C (18 F) and 19 °C (34 F), respectively, approach to the adiabatic saturation temperature. Next, the impact of the calcium stoichiometry and the inlet concentration of  $\text{SO}_2$  were investigated. Based on the result from the bench scale experiments, a set of operating conditions was selected for the pilot plant. The test runs in the pilot plant were performed over a longer period of time in order to closer study the impact of variations of the pressure drop over the fabric filter. Both lime and limestone were utilized.

## EXPERIMENTAL RESULTS

It was clear from the first tests in the bench scale spray dry scrubber that addition of sodium chlorite to a lime slurry enables a substantial amount of  $\text{NO}_x$  to be removed. This is displayed in Figure 3, which presents data from experiments at 40 and 62 percent relative humidity. In both cases the inlet concentrations were set at 1000 ppm  $\text{SO}_2$  and 300 ppm  $\text{NO}_x$ . As can be seen, an optimum is indicated with respect to the amount of sodium chlorite added. The optimum appears to be at a molar ratio  $\text{NaClO}_2/\text{SO}_2$  (additive ratio) of about 0.6, corresponding to a molar ratio  $\text{NaClO}_2/\text{NO} = 2$ . The impact of the relative humidity or the approach to the saturation point seems to be of minor importance for the removal of  $\text{NO}_x$ . Up to 75 percent removal was obtained at the highest relative humidity and optimal amount of additive.

It can also be seen from the figure that sodium chlorite has a tremendous impact on the removal of  $\text{SO}_2$ . At an additive ratio as low as 0.2, greater than 95 percent  $\text{SO}_2$  removal can be achieved. The enhancement of the sulfur capture may be due to the oxidation of sulfite to sulfate, in which case hydrogen ions are released from bisulfite. This increases the solubility of  $\text{SO}_2$ . However, this cannot be the only explanation, because there is only chlorite available for oxidation of about 15 percent of the sulfur at an additive ratio of 0.2. Several other effects may be of significance, including interactions between species of nitrogen and sulfur, as well as enhancing effects from the production of chloride ion [18]. The former effect is known from simultaneous wet scrubbing of  $\text{NO}_x$  and  $\text{SO}_2$ . Furthermore, it is clear from the figure that smaller amounts of chlorite are needed for high  $\text{SO}_2$  removal at higher relative humidities.

Figure 4 shows the results from tests with limestone. The operating conditions were set similar to those used in the lime case. As can be seen, the behaviour of  $\text{NO}_x$  in the limestone case is similar to that for the lime case. However, the optimum is more pronounced in the limestone case, with 75 percent removal at an additive ratio of 0.6 and a relative humidity of 62 percent. Surprisingly high removal efficiencies of  $\text{SO}_2$  were seen in the limestone case. In fact, additive and limestone removed as much  $\text{SO}_2$  at the optimal additive ratio as an unpromoted lime system. Hence, a limestone system has the potential to be used for the removal of both  $\text{SO}_2$  and  $\text{NO}_x$ .

When considering the different  $\text{NO}_x$  species present in the system, it can be seen from Figures 5 and 6 that NO is oxidized to  $\text{NO}_2$  concurrent with the absorption of NO. The  $\text{NO}_x$  exiting the system contains a larger fraction of  $\text{NO}_2$  at higher removal efficiencies. At an additive ratio of 0.5, the  $\text{NO}_x$  leaving the fabric filter consists of about equal amounts of NO and  $\text{NO}_2$ . Furthermore, it appears that some of the  $\text{NO}_2$  produced in the spray dryer is reduced back to NO in the fabric filter; even though the  $\text{NO}_x$  concentration drops over the fabric filter, the NO concentration increases somewhat. This situation can only occur if the dry end product is completely exhausted with respect to chlorite and alkali. If no chlorite is present, reducing conditions may prevail. When analyzing the product from the tests, not even



traces of chlorite could be detected. Lack of alkali is required, because the reduction reaction proceeds over nitrous acid as an intermediate; nitrous acid decomposes to NO. If alkali is present, nitrous acid is stabilized due to the formation of nitrite ion. The analyses showed no alkali, which should be anticipated from Figure 3. In fact, to obtain 75 percent NO<sub>x</sub> removal and 97 percent SO<sub>2</sub> removal, a stoichiometric ratio of 1.08 is required. However, the set stoichiometry was 1.0. This discrepancy can only be explained by inaccuracies in the determination of the feeding rate of the lime slurry.

There are two possible mechanisms for the oxidation of NO to NO<sub>2</sub>. First, NO may be oxidized by chlorite in the liquid phase, with subsequent desorption of NO<sub>2</sub>. Second, chlorite may decompose to chlorine dioxide, which desorbs from the droplets; the chlorine dioxide oxidizes the NO in the gas phase.

Further tests were done in the bench scale equipment to outline the impact of various parameters. Data from these tests have all been presented in terms of the overall removal efficiency, thus excluding the removal in the spray dryer. First, the impact of the stoichiometric ratio was studied. In all these tests, the additive ratio (NaClO<sub>2</sub>/SO<sub>2</sub>) was set at 0.5, which is somewhat lower than the optimal value with respect to the NO<sub>x</sub> removal. It was seen quite clear that the stoichiometry has a negligible impact when it exceeds one. However, the data have been included to enable a closer comparison between lime and limestone, Figures 7 and 8. When considering NO, it can be seen from the figures that the relative humidity has very little impact at the actual additive ratio. Furthermore, lime and limestone show similar behaviour. The only difference can be obtained for SO<sub>2</sub> at stoichiometries close to unity, in which case lime is superior. However, utilizing excess of limestone leads to a tremendous removal of SO<sub>2</sub>.

Figures 9 through 12 show bench scale data from different SO<sub>2</sub> to NO<sub>x</sub> ratios at the inlet. The diagrams all display the dependency of the concentration of SO<sub>2</sub> at 300 ppm NO<sub>x</sub> at the inlet on the removal efficiency. Even though data for lime and limestone are not completely similar, the trends obtained for the two absorbents are identical. The interpretation of the result is quite complicated. There are actually four different reagent ratios of interest, ie. Ca/S, Ca/NO<sub>x</sub>, Na/S and Na/NO<sub>x</sub>. It is obvious from the diagrams that the Na/NO<sub>x</sub> ratio is of utmost importance for the removal of NO<sub>x</sub>; the removal decreases if the ratio mentioned is decreased according to Figures 9 and 11. The behaviour of SO<sub>2</sub> is unexpected; as can be seen from Figures 10 and 12, the removal decreases when the Na/S ratio is increased due to the decrease in SO<sub>2</sub> concentration. This is true both when the Ca/S ratio is kept constant and when it increases.

Test runs in the pilot plant were undertaken to verify the results on a larger scale (500 x). The results from these test runs are summarized in Figures 13 through 15. The impact of the pressure drop over the fabric filter was investigated in the range from 0.3 kPa (pressure drop over cloth) up to 2.0 kPa. Only very small variations of the removal efficiency were observed when varying the pressure drop. This holds for both SO<sub>2</sub> and NO<sub>x</sub>. The further experiments were steady state runs at a high pressure drop.

Figures 13 and 14 are test runs with lime and limestone, respectively, at a stoichiometric ratio (Ca/S) of about 1.2 and an additive ratio (Na/S) of 0.5. This is somewhat below the point for the optimal amount of additive. The results generally verify the bench scale tests. This is especially true for NO<sub>x</sub> removal. However, the removal of SO<sub>2</sub> is somewhat lower, compared to the bench scale tests. By increasing the stoichiometry to 1.4, the removal of both species increases, as shown in Figure 15. The data show up to 70 percent removal of NO<sub>x</sub>.

## DISCUSSION

### END PRODUCT PROPERTIES AND COMPOSITION

Samples of dry end product were continuously taken during the bench scale and the pilot plant runs. This includes dropout from the spray dryer as well as samples taken from the filter cloth. The product displayed very desirable properties. The solids were free flowing and possessed a high degree of flightability. Furthermore, the product was easily dried in the spray dryer. The content of free moisture in 15 product samples from the pilot plant was found to range from 0.0 to 2.7 percent, with an average of 0.7 percent; the free moisture has here been defined as sample weight loss when heated to 150 °C.

The product was shown to have a low ability to stick to surfaces. A high degree of stickiness is considered a problem in spray dry scrubbing, because of the potential for buildup of soft scale. The formation of soft scale on the spray nozzle was inspected visually. It was clearly seen that sodium chloride as an additive decreased the tendency for soft scale buildup. In fact, the tendency for soft scale buildup on the spray nozzle was found to decrease according to the following series:

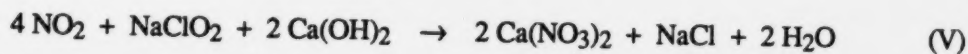
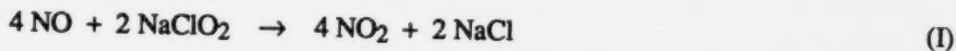
Lime > Limestone > Lime/Additive > Limestone/Additive

The product composition was analyzed according to different methods. Major constituents were found to be sodium chloride (NaCl), calcium nitrate ( $\text{Ca}(\text{NO}_3)_2$ ), calcium sulfate ( $\text{Ca}(\text{SO}_4)$ ) and unreacted alkali ( $\text{Ca}(\text{OH})_2/\text{CaCO}_3$ ). Cations were analyzed by atomic absorption and anions were analyzed by ion chromatography. In addition, sulfite was analyzed by iodometric titration and the total sulfur content was determined on a Leco sulfur analyzer. From the analyses of a large number of samples it was found that the recovery ratio of the different species was good, when considering the problems with taking representative samples as well as sample preparation. All the samples from the bench scale tests were shown to be free from anions such as nitrite, sulfite, chlorate etc.; not even traces of these species were detected. However, the samples from the pilot tests were shown to contain small traces of nitrite, sulfite and unreacted chlorite; no other species were identified.

In conclusion, the end product should be anticipated to have excellent properties and well defined composition. No sulfite will be present and all sulfur will be in the form of calcium sulfate. All chlorite is converted to chloride and nitrogen is recovered as nitrate and not nitrite. However, the sodium chloride and the calcium nitrate are highly soluble. Considering the operating conditions of Figure 15 at 60 percent relative humidity, the anticipated product composition is in weight percent : 64 %  $\text{Ca}(\text{SO}_4)$ , 14 %  $\text{CaCO}_3$ , 14 % NaCl and 8 %  $\text{Ca}(\text{NO}_3)_2$ . Studies of options for the recovery or disposal of the end product have not yet been undertaken.

#### CHEMISTRY AND MECHANISMS

The mechanism for absorption of  $\text{NO}_x$  is believed to be a two step reaction. Sodium chlorite does not oxidize NO directly to nitrate. The reaction product is either  $\text{NO}_2$  or nitrite. The further oxidation of nitrite is known to be very slow [21], and could be neglected in a spray dry system. Furthermore, the dry end product contained no or only small traces of nitrite. Hence, NO is first oxidized to  $\text{NO}_2$ , which is further oxidized to nitrate. The first of these reactions is also apparent from the formation of  $\text{NO}_2$  measured in the exit gas. The two step mechanism would then be:



The rate of these reactions depends on the chlorite concentration as well as the pH value in a complex way. The fact that an optimal additive ratio exists can be explained by the change in the ratio of the reaction rates. To prevent the  $\text{NO}_2$  from desorbing from the droplets, reaction (V) must be several orders of magnitude faster than reaction (I), which is not the case under any conditions. To prevent the formation of  $\text{NO}_2$ , the pH must be extremely high. However, this leads to the formation of nitrite in the end product.

There are several indications that gas phase reactions may play an important role in the absorption of  $\text{NO}_x$ . One possible gas phase mechanism starts with the decomposition of chlorite to chlorine dioxide. Chlorine dioxide desorbs from the liquid phase to oxidize both  $\text{SO}_2$  and NO in the gas phase. The first indication of gas phase reactions is the theoretical outline for reaction kinetics. It was shown that the reaction rates required for the reactions in the droplets are much faster than those measured in small scale laboratory experiments. Second, the removal efficiencies measured for  $\text{SO}_2$  appear to be much higher than the theoretically highest possible values [16]. This indicates the involvement of a gas phase mechanism to assist the absorption of  $\text{SO}_2$ . Third, a small but significant discrepancy was seen when comparing the bench scale tests with the pilot test runs. This may be an indication of gas phase reactions, because the two devices do not usually show any scale effects. Fourth, many of the parameters investigated, such as the relative humidity and the stoichiometric ratio, did not show the significant

impact on the NO<sub>x</sub> removal as is seen in conventional spray dry scrubbing systems. However, the additive ratio was shown to be very important.

It is obviously an interest to investigate if chlorine dioxide (ClO<sub>2</sub>) is present in the gas phase, because emission of this species is undesirable. Hence, manual gas samples were continuously taken during the test runs to determine ClO<sub>2</sub> by absorption in bottle trains. However, it is very difficult to determine ClO<sub>2</sub> in the presence of SO<sub>2</sub>, because these species react in a similar fashion with a number of absorbents. The concentration of ClO<sub>2</sub> must then be estimated as a difference. This gives unreliable results if the concentration of SO<sub>2</sub> is much higher than the concentration of ClO<sub>2</sub>. Therefore, reliable analyses can only be done when the removal efficiency of SO<sub>2</sub> is virtually 100 percent in the spray dry scrubbing system. This also occurred several times. The chlorine dioxide was absorbed in a potassium iodine solution in the bottle train and the determination of total SO<sub>2</sub> and ClO<sub>2</sub> was done by iodometric titration. The chlorine dioxide was determined by difference. These analyses showed that the concentration of chlorine dioxide was 5 ppm or less after the fabric filter. The analyses of the end product showed that the recovery ratio of chlorine was 100 percent, within the accuracies provided by the analysis procedures. This obviously indicates that the concentration of chlorine dioxide in the gas phase must be very low.

Even though all sulfur was found to be oxidized to sulfate in the end product, sodium chlorite was only responsible for oxidizing part of the sulfite; this oxidation would be according to reaction (IV). This was found because less than the theoretical amount of sodium chlorite was consumed for oxidation of sulfur and nitrogen species; i.e. all sulfur was in the form of sulfate even at very low additive ratios. Hence, the oxygen of the flue gas must be responsible for a substantial part of the sulfite oxidation. For example, chloride ion from consumed chlorite enhances this reaction. According to previous experiments chloride increased the percentage of sulfite oxidation in a limestone based system from 40 percent in the absence of chloride to almost 60 percent when chloride is added. Lime gave 15 percent sulfate under the same conditions. It is also known from previous studies that NO<sub>2</sub> has a catalytic effect on the oxidation of sulfite [22].

## ECONOMICS

The cost for the modification of a conventionally designed spray dry scrubber system to enable NO<sub>x</sub> control can be divided into three cost items; i.e. the cost for constructional modifications, the cost for more cumbersome procedures for disposal or recovery of the end product, and the cost for the additive. In addition, if a limestone based system is selected, credit for changing from lime to limestone must be subtracted.

The constructional modifications are anticipated to be minor. The mechanical product properties are in fact improved when utilizing an additive. The additional equipment required includes holding tanks and/or storage silos for the additive etc.

The cost introduced because of the change in amounts of soluble species in the end product may or may not be substantial. The actual cost will depend on local legislation. As previously mentioned, methods for disposal and recovery of the product have not been assessed.

The major cost item is represented by the cost of the additive. Based on current market prices, this cost item is extremely high. Worldwide, the price of sodium chlorite is of the order of US\$ 2,000 - 3,000 per ton, estimated at 100 % NaClO<sub>2</sub>. There are two reasons for this high price. First, the production cost is high because expensive reactants are utilized in present production processes. Second, the production capacity is very small on a worldwide basis. There are a handful of producers in Europe, a few in Japan and only a single producer in the United States. The total production capacity within the European Common Market is of the order 20,000 tons annually. To put this figure in perspective, it should be compared with required consumption rates when utilized as an additive in a spray dry scrubbing process. In fact, the 20,000 tons will only be sufficient to cover the demand for a single spray dry scrubbing system treating the flue gas from a 250 MW utility boiler. Clearly, the actual cost for producing sodium chlorite on a large scale has to be assessed.

An estimate based on current market prices of sodium chlorite indicates a cost of about 20-30 mills/kWh for 70 percent NO<sub>x</sub> control by spray dry scrubbing. The leading control technology for



efficient NO<sub>x</sub> control on utility boilers is presently selective catalytic reduction (SCR) with ammonia. The cost for 80 percent NO<sub>x</sub> control with SCR is claimed to be 5-15 mills/kWh.

## CONCLUSIONS

The removal of NO<sub>x</sub> concurrently with SO<sub>2</sub> was studied in spray dry scrubbers on the bench scale as well as on the pilot plant scale. Slurries were prepared from lime and limestone using sodium chlorite as an additive. Up to 75 percent removal efficiency of NO<sub>x</sub> was obtained for both lime and limestone. However, uncaptured NO<sub>x</sub> will partly exist in the form of NO<sub>2</sub>. Furthermore, the removal of SO<sub>2</sub> increased due to the additive.

The end product has good mechanical properties, and the major product species are calcium sulfate, calcium nitrate, sodium chloride and unreacted alkali. Methods for treating and disposal of the product have not been investigated. A process based on sodium chlorite is very expensive, if estimates are based on present market prices of sodium chlorite.

## ACKNOWLEDGEMENT

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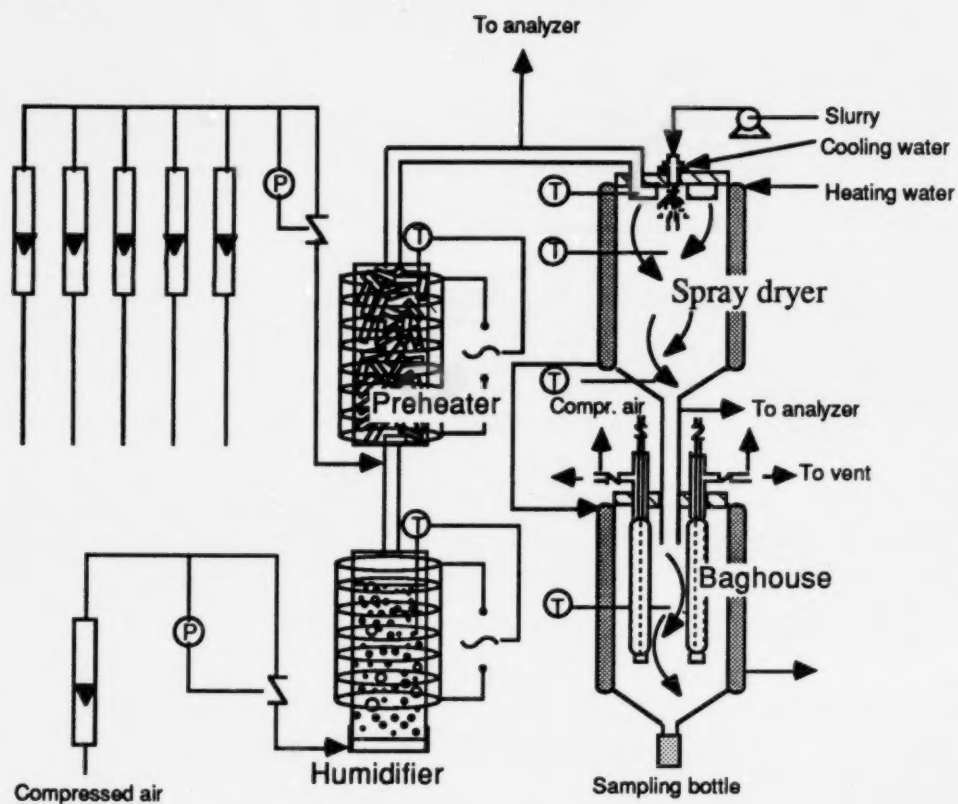


Figure 1. The bench scale spray dry scrubber

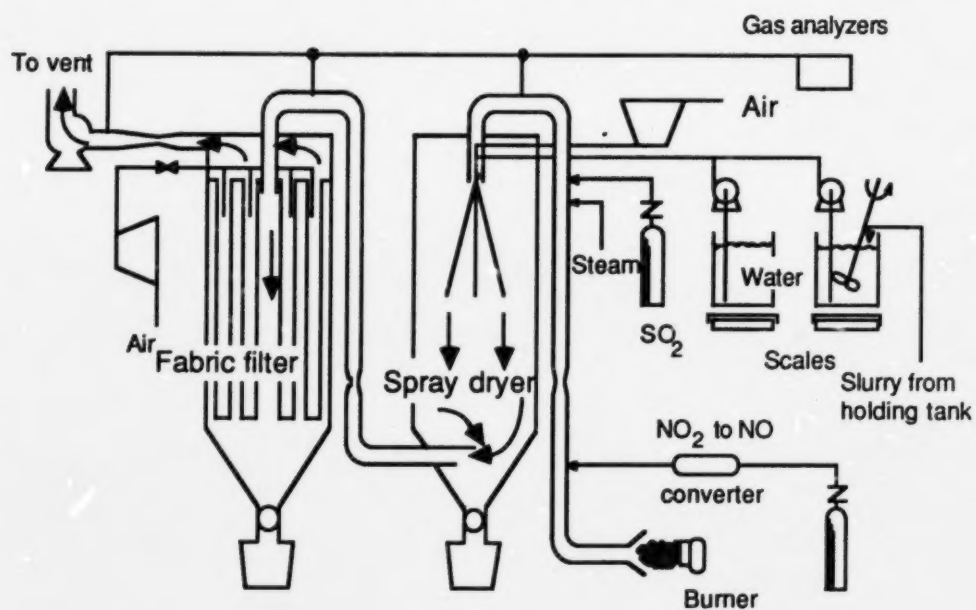


Figure 2. The pilot plant

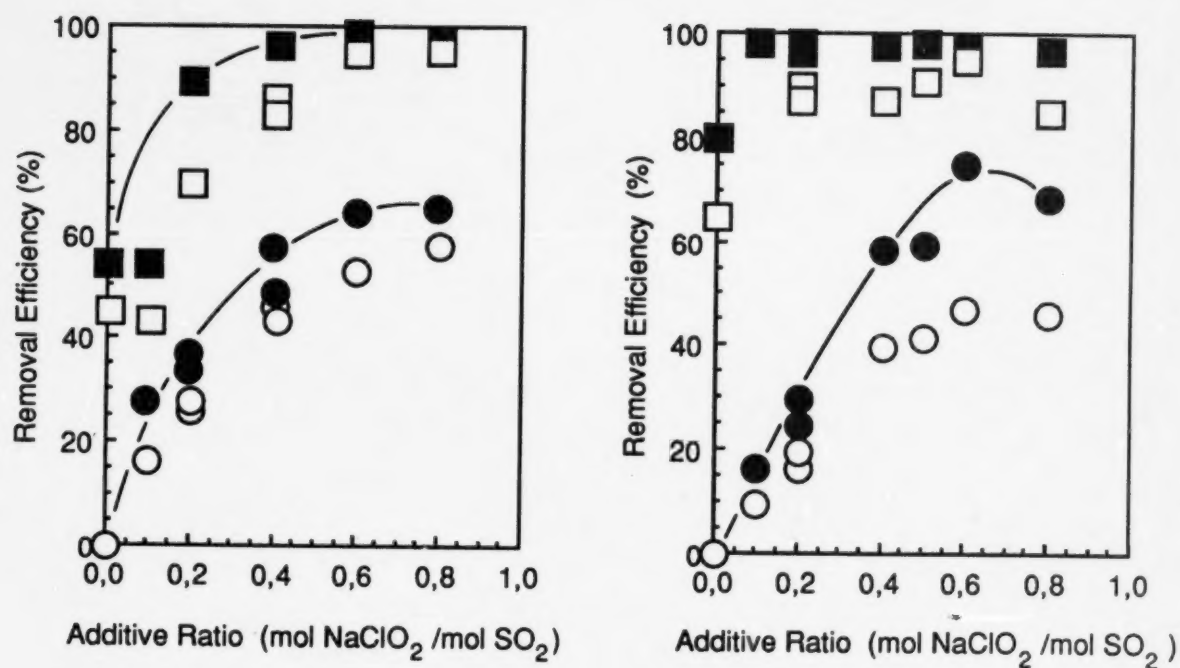


Figure 3. Removal efficiency with lime at a stoichiometric ratio (Ca/S) of one; 40 % (left) and 62 % (right) relative humidity; circles are NO<sub>x</sub> and squares are SO<sub>2</sub>; open symbols are spray dryer removal and filled symbols are overall removal; 1000 ppm SO<sub>2</sub> and 300 ppm NO<sub>x</sub> at inlet

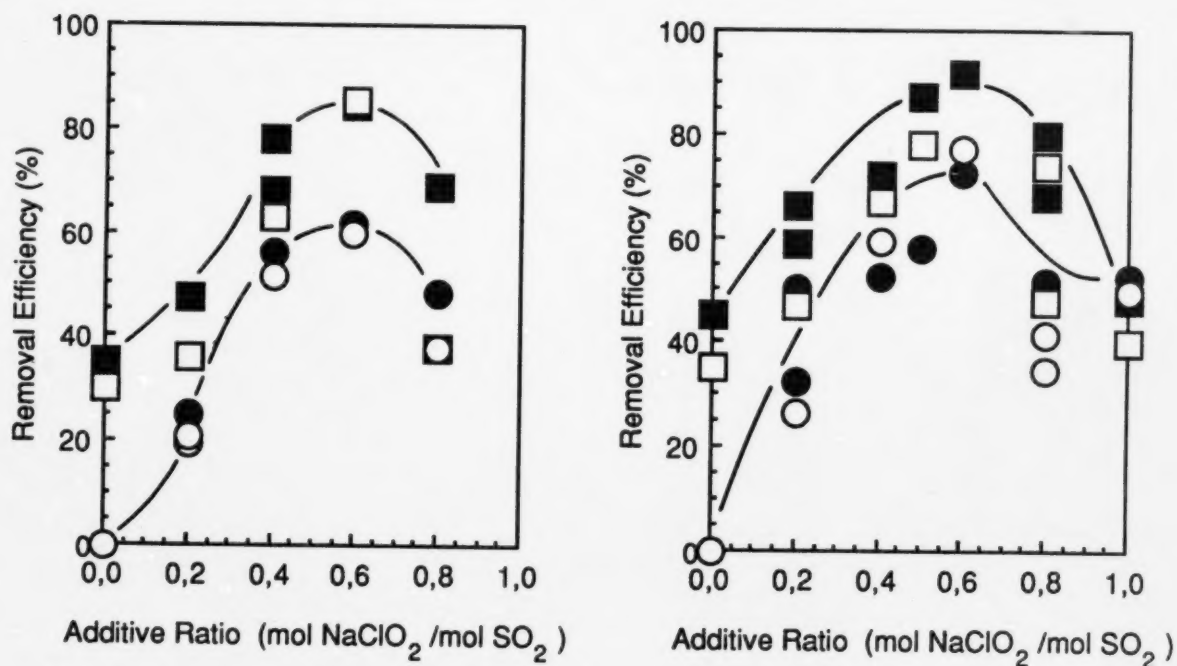


Figure 4. Removal efficiency with limestone at a stoichiometric ratio (Ca/S) of one; 40 % (left) and 62 % (right) relative humidity; circles are NO<sub>x</sub> and squares are SO<sub>2</sub>; open symbols are spray dryer removal and filled symbols are overall removal; 1000 ppm SO<sub>2</sub> and 300 ppm NO<sub>x</sub> at inlet

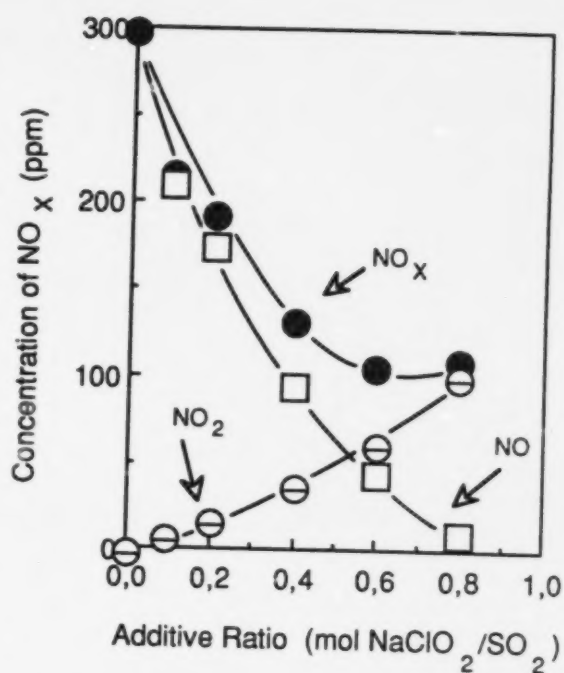
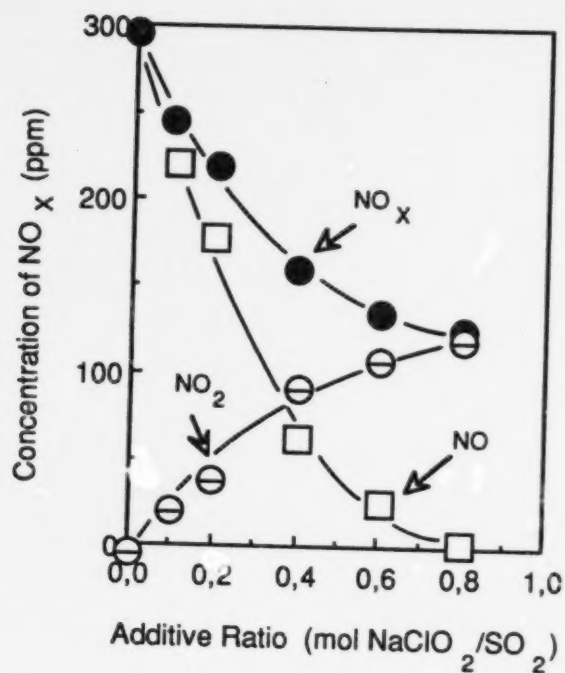


Figure 5. Concentration of NO<sub>x</sub> species measured after the spray dryer (left) and after the fabric filter (right), respectively. Data for lime slurry at 40 % relative humidity (19 °C/34 F approach to the saturation temperature) and 1000 ppm SO<sub>2</sub> at inlet

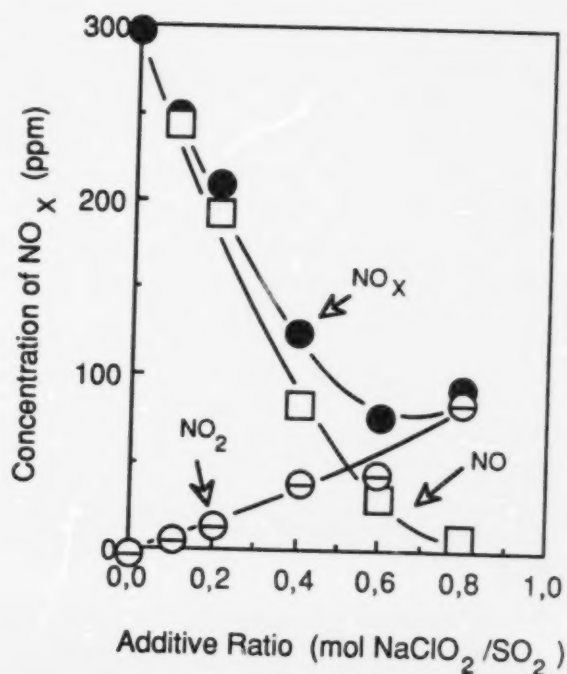
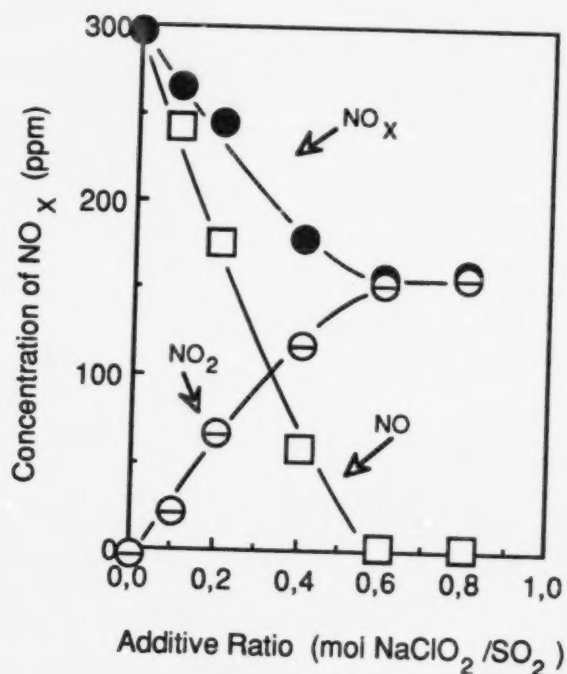


Figure 6. Concentration of NO<sub>x</sub> species measured after the spray dryer (left) and after the fabric filter (right), respectively. Data for lime slurry at 62 % relative humidity (9 °C/16 F approach to the saturation temperature) and 1000 ppm SO<sub>2</sub> at inlet



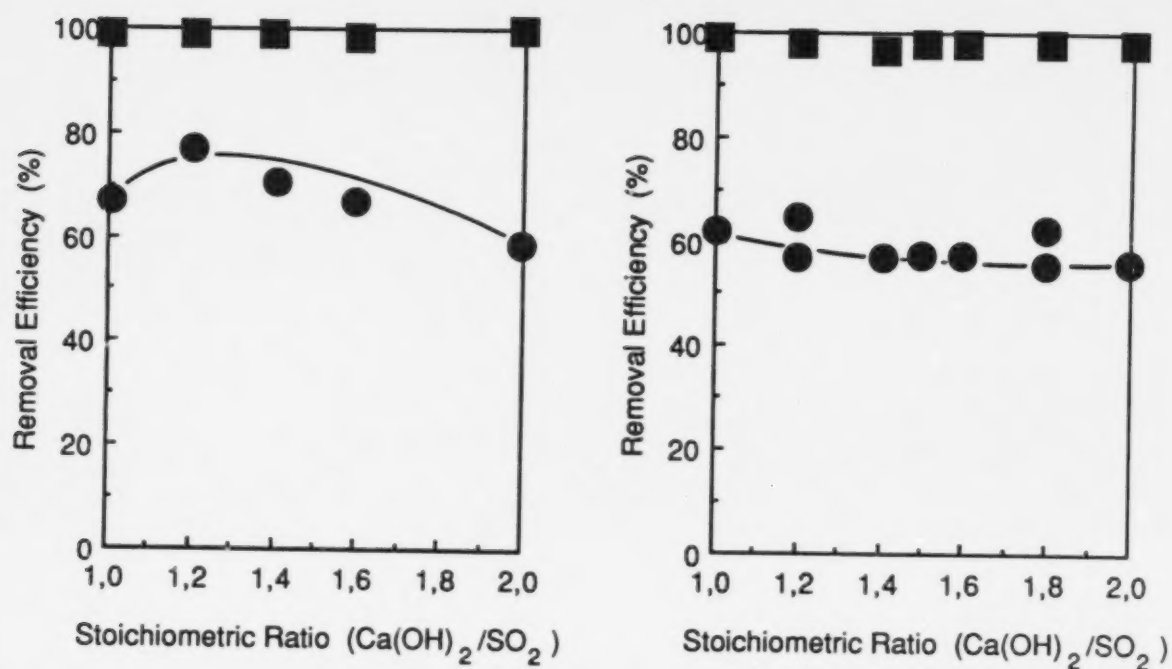


Figure 7. Overall removal efficiency with lime slurry at an additive ratio (Na/S) of 0.5; 40 % (left) and 62 % (right) relative humidity; circles are  $\text{NO}_x$  and squares are  $\text{SO}_2$ ; 1000 ppm  $\text{SO}_2$  and 300 ppm  $\text{NO}_x$  at inlet

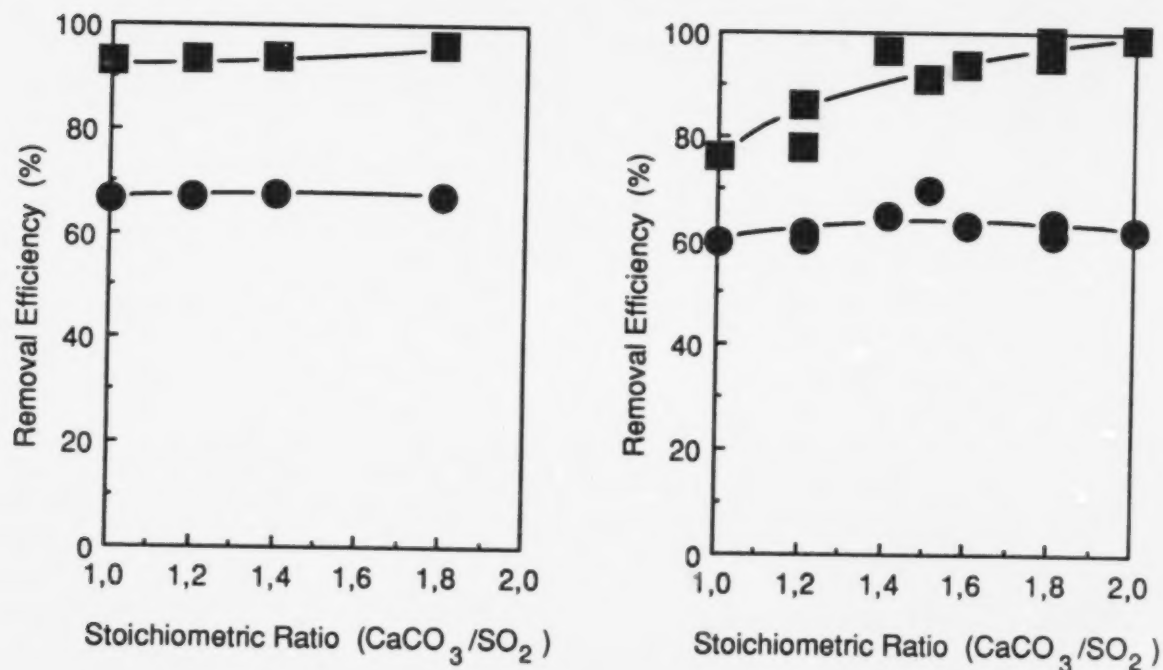


Figure 8. Overall removal efficiency with limestone at an additive ratio (Na/S) of 0.5; 40 % (left) and 62 % (right) relative humidity; circles are  $\text{NO}_x$  and squares are  $\text{SO}_2$ ; 1000 ppm  $\text{SO}_2$  and 300 ppm  $\text{NO}_x$  at inlet

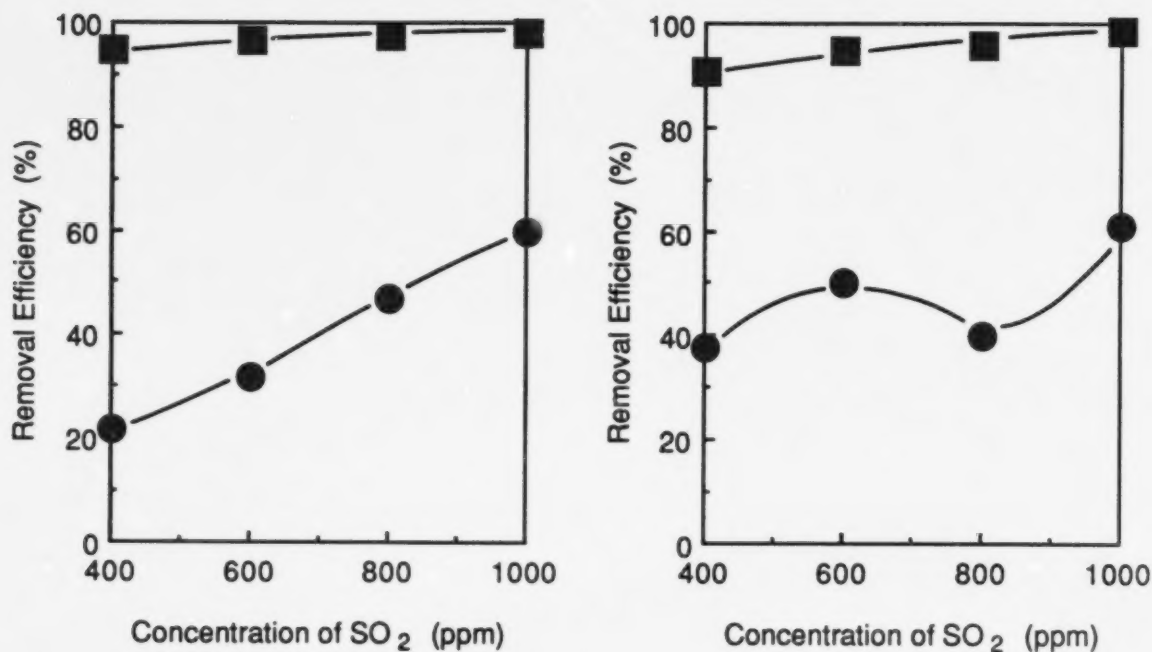


Figure 9. Overall removal efficiency of SO<sub>2</sub> (squares) and NO<sub>x</sub> (circles) with lime at an additive ratio (NaClO<sub>2</sub>/SO<sub>2</sub>) of 0.5; 300 ppm NO<sub>x</sub> at inlet and 62 % relative humidity. Left: Ca/S = 1; Ca/NO<sub>x</sub> and Na/NO<sub>x</sub> vary. Right: Ca/NO<sub>x</sub> = 3.3; Na/NO<sub>x</sub> and Ca/S vary.

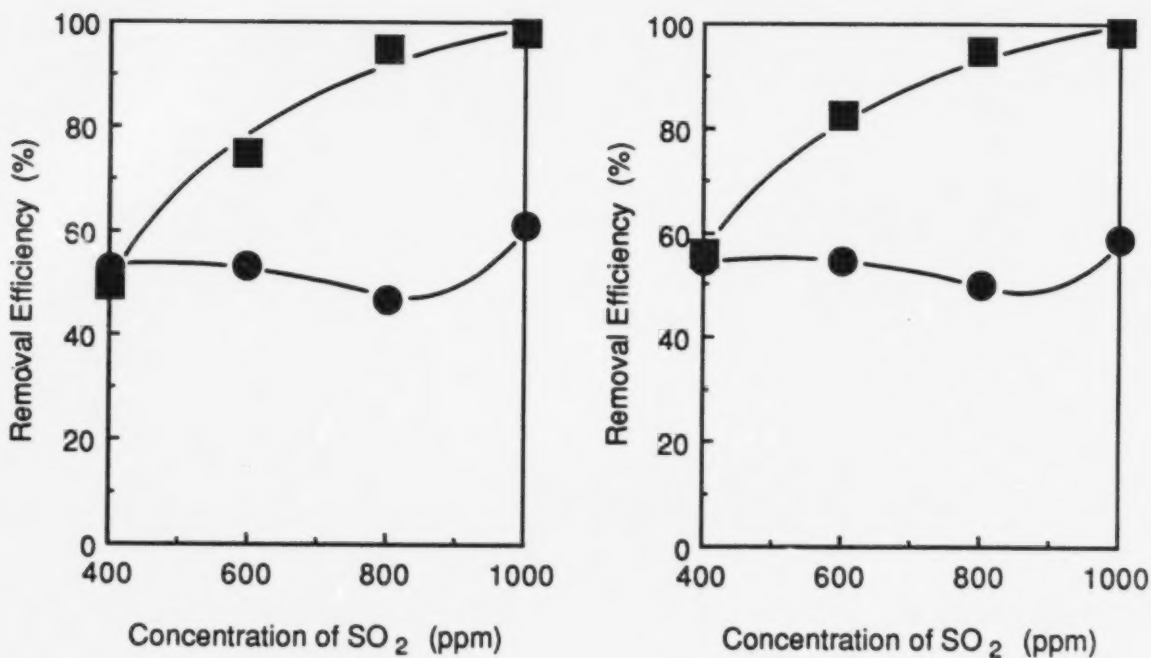


Figure 10. Overall removal efficiency of SO<sub>2</sub> (squares) and NO<sub>x</sub> (circles) with lime at an additive ratio (NaClO<sub>2</sub>/NO<sub>x</sub>) of 1.67; 300 ppm NO<sub>x</sub> at inlet and 62 % relative humidity. Left: Ca/S = 1; Ca/NO<sub>x</sub> and Na/S vary. Right: Ca/NO<sub>x</sub> = 3.3; Na/S and Ca/S vary.

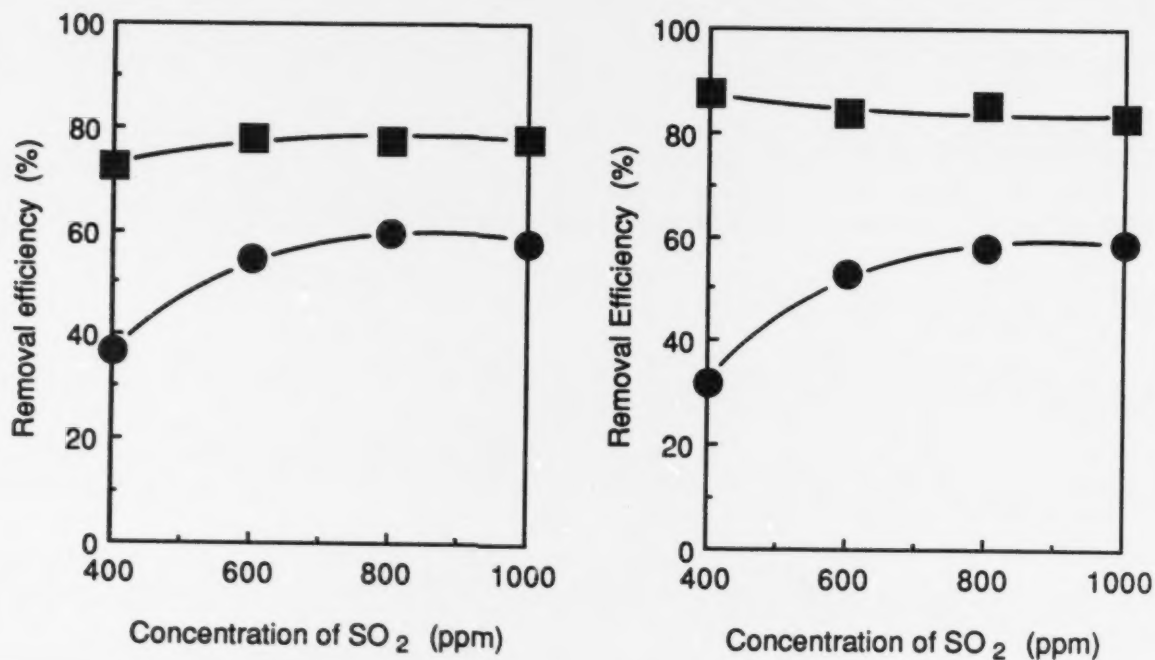


Figure 11. Overall removal efficiency of SO<sub>2</sub> (squares) and NO<sub>x</sub> (circles) with limestone at an additive ratio (NaClO<sub>2</sub>/SO<sub>2</sub>) of 0.5; 300 ppm NO<sub>x</sub> at inlet and 62 % relative humidity. Left: Ca/S = 1; Ca/NO<sub>x</sub> and Na/NO<sub>x</sub> vary. Right: Ca/NO<sub>x</sub> = 3.3; Na/NO<sub>x</sub> and Ca/S vary.

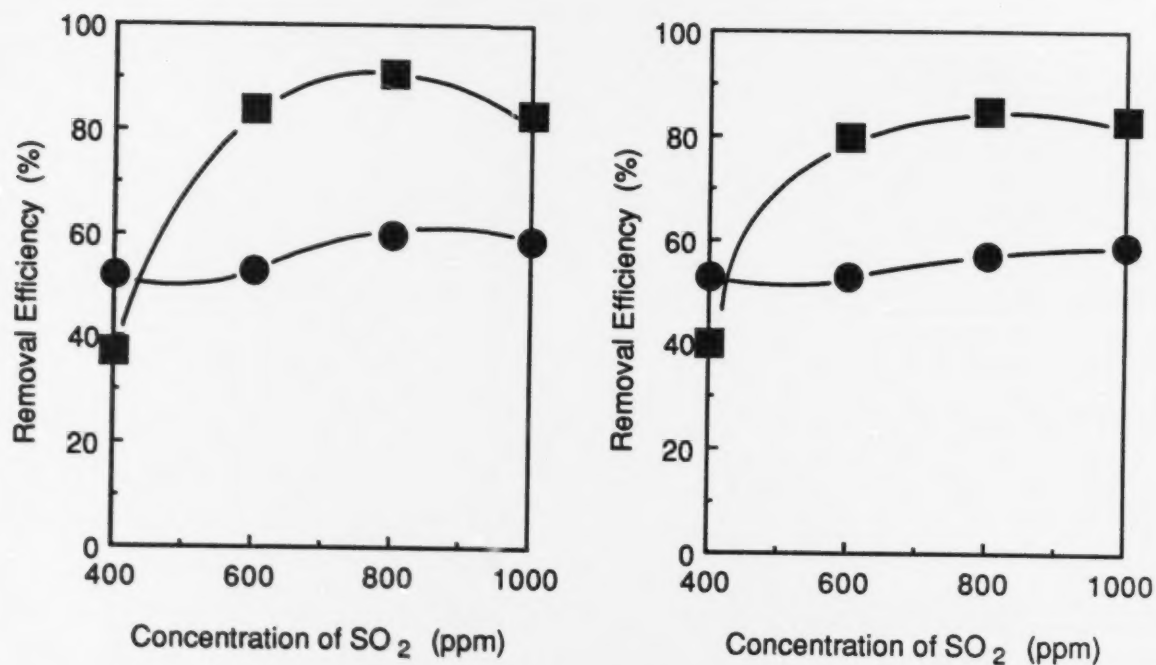


Figure 12. Overall removal efficiency of SO<sub>2</sub> (squares) and NO<sub>x</sub> (circles) with limestone at an additive ratio (NaClO<sub>2</sub>/NO<sub>x</sub>) of 1.67; 300 ppm NO<sub>x</sub> at inlet and 62 % relative humidity. Left: Ca/S = 1; Ca/NO<sub>x</sub> and Na/S vary. Right: Ca/NO<sub>x</sub> = 3.3; Na/S and Ca/S vary.

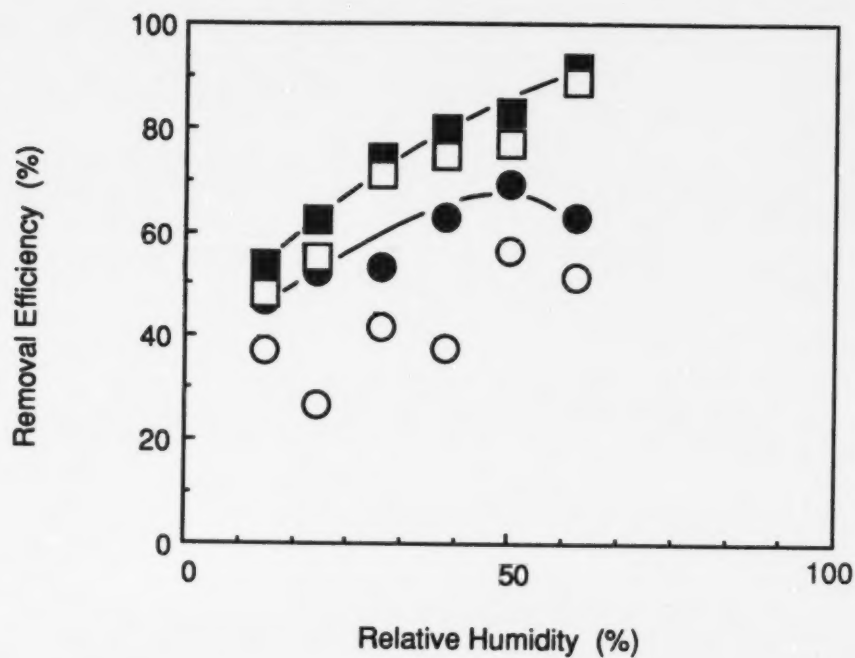


Figure 13. Pilot plant removal - spray dryer (open) and overall (filled) - with lime at an additive ratio ( $\text{NaClO}_2/\text{SO}_2$ ) of 0.5 and a stoichiometric ratio ( $\text{Ca}/\text{S}$ ) of 1.2; 300-350 ppm  $\text{NO}_x$ ;  $\text{SO}_2$  (squares) and  $\text{NO}_x$  (circles)

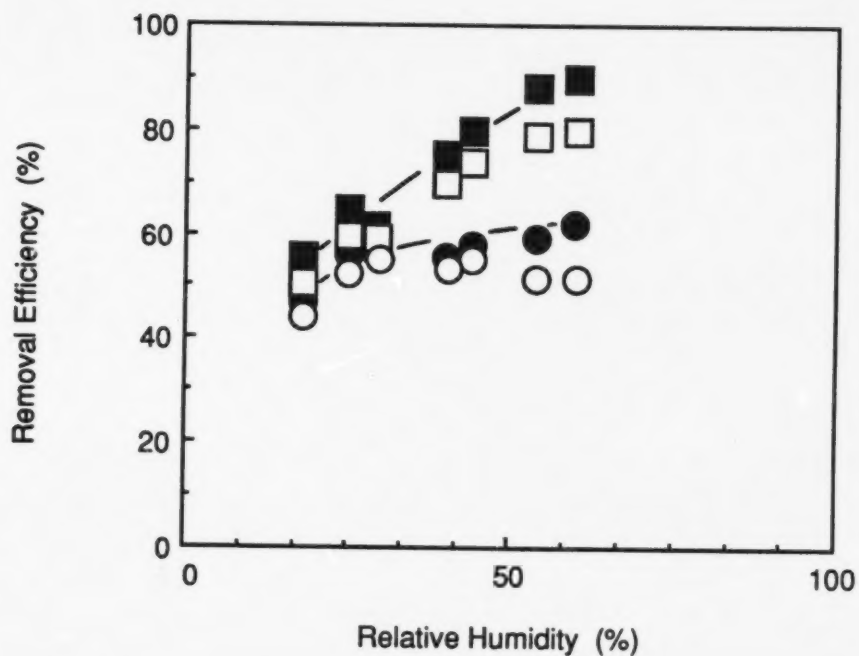


Figure 14. Pilot plant removal - spray dryer (open) and overall (filled) - with limestone at an additive ratio ( $\text{NaClO}_2/\text{SO}_2$ ) of 0.5 and a stoichiometric ratio ( $\text{Ca}/\text{S}$ ) of 1.2; 300-350 ppm  $\text{NO}_x$ ;  $\text{SO}_2$  (squares) and  $\text{NO}_x$  (circles)



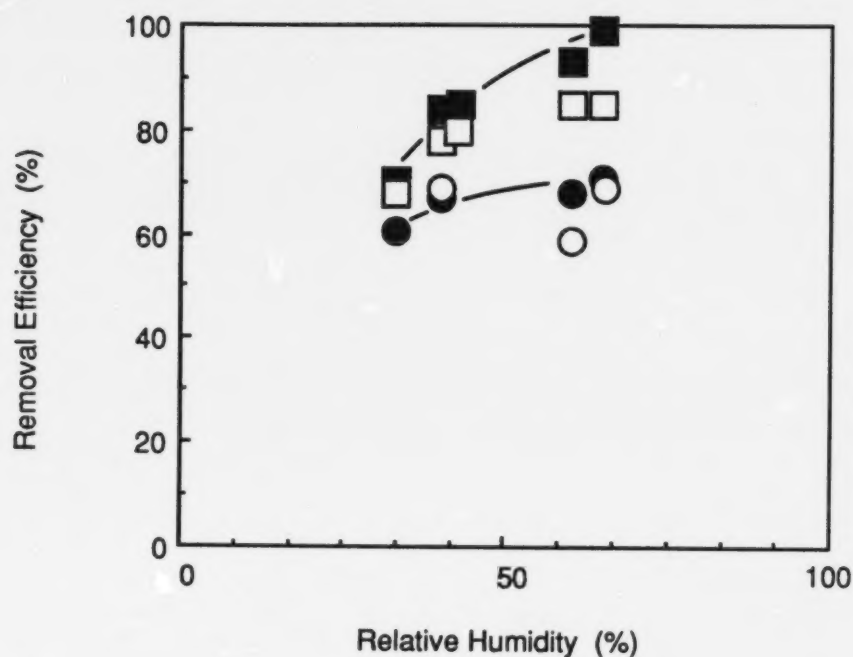


Figure 15. Pilot plant removal - spray dryer (open) and overall (filled) - with limestone at an additive ratio ( $\text{NaClO}_2/\text{SO}_2$ ) of 0.5 and a stoichiometric ratio ( $\text{Ca/S}$ ) of 1.4; 300-350 ppm  $\text{NO}_x$ ;  $\text{SO}_2$  (squares) and  $\text{NO}_x$  (circles)

TABLE 1. SAND BED TESTS WITH ADDITIVE AND  $\text{Ca}(\text{OH})_2$ ; +, -, AND 0 INDICATE INCREASED, DECREASED AND UNCHANGED EFFICIENCY, RESPECTIVELY, WHEN COMPARED TO PURE LIME

| Additive                                                      | NO | NO <sub>2</sub> | SO <sub>2</sub> | Additive                                                                  | NO | NO <sub>2</sub> | SO <sub>2</sub> |
|---------------------------------------------------------------|----|-----------------|-----------------|---------------------------------------------------------------------------|----|-----------------|-----------------|
| $\text{Al}_2\text{O}_3$                                       | +  | -               | +               | $\text{Fe}_2\text{O}_3/\text{Al}_2\text{O}_3$                             | -  | 0               | -               |
| $\text{Na}_2\text{CO}_3/\text{Al}_2\text{O}_3$                | +  | -               | +               | $\text{CH}_3\text{COONH}_4$                                               | -  | -               | +               |
| $\text{Cr}_2\text{O}_3$                                       | +  | -               | +               | Cu                                                                        | +  | -               | +               |
| CuO                                                           | -  | +               | -               | $\text{Al}_2\text{O}_3/\text{CuO}$                                        | -  | -               | -               |
| $\text{Cu}_2\text{O}$                                         | +  | 0               | +               | EDTA                                                                      | -  | -               | -               |
| $\text{Fe}_2\text{O}_3/\text{EDTA}$                           | -  | +               | -               | $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$                      | -  | +               | +               |
| $\text{Fe}_2\text{O}_3$                                       | -  | +               | -               | $\text{Fe}_2\text{O}_3/\text{MnO}/\text{ZnO}$                             | -  | -               | 0               |
| $\text{Fe}_2\text{O}_3/\text{Al}_2\text{O}_3/\text{MoO}_3$    | -  | +               | -               | $\text{FeS}/\text{CuCl}$                                                  | -  | +               | +               |
| $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$                     | -  | -               | -               | $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$                                 | -  | -               | -               |
| $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$                     | -  | -               | -               | $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$                      | 0  | 0               | +               |
| KOH                                                           | -  | -               | -               | $\text{Mg}(\text{OH})_2$                                                  | +  | 0               | +               |
| $\text{MnO}/\text{Al}_2\text{O}_3$                            | +  | -               | +               | $\text{C}_{19}\text{H}_{42}\text{BrN}$                                    | -  | +               | +               |
| $\text{C}_{19}\text{H}_{42}\text{BrN}/\text{Urea}$            | -  | +               | +               | $\text{C}_{19}\text{H}_{42}\text{BrN}/\text{Urea}/\text{Na}_2\text{SO}_3$ | -  | +               | +               |
| NiO                                                           | +  | -               | +               | $\text{KIO}_4$                                                            | +  | +               | +               |
| $\text{C}_4\text{H}_4\text{KNaO}_6 \cdot 4\text{H}_2\text{O}$ | -  | +               | -               | NaOH                                                                      | +  | -               | +               |
| $\text{NaNO}_2$                                               | -  | -               | -               | $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$                       | +  | -               | +               |
| $\text{Na}_2\text{SO}_3 \cdot 7\text{H}_2\text{O}$            | +  | -               | +               | Urea                                                                      | +  | -               | +               |
| $\text{Zn}(\text{OH})_2$                                      | 0  | -               | 0               | ZnO                                                                       | -  | +               | -               |
| Urea/NaOH                                                     | +  | +               | +               | $\text{NaAlO}_2$                                                          | 0  | +               | +               |
| $\text{KMnO}_4$                                               | +  | +               | +               | $\text{NaClO}_2$                                                          | +  | +               | +               |

TABLE 2. DESIGN AND OPERATING CONDITIONS FOR  
SPRAY DRYER-BAGHOUSE SYSTEMS

|                                             | Bench scale            | Pilot plant            |
|---------------------------------------------|------------------------|------------------------|
| Spray dryer                                 |                        |                        |
| Nozzle type:                                | Ultrasonic             | Dual media             |
| Diameter:                                   | 0.1 m                  | 1.2 m                  |
| Total height                                | 0.6 m                  | 5.0 m                  |
| Baghouse                                    |                        |                        |
| Diameter:                                   | 0.1m                   | 1.2 m                  |
| Total height:                               | 0.35 m                 | 5.0 m                  |
| Bag design:                                 | high ratio/jet pulse   |                        |
| Total number of bags:                       | 5                      | 20                     |
| Total filter area:                          | 0.062 m <sup>2</sup>   | 11.2m <sup>2</sup>     |
| Type of fabric:                             | needle type/polyamide  |                        |
| Experimental conditions                     |                        |                        |
| Concentration of SO <sub>2</sub> (dry gas): | 400-1000 ppm           | 1000 ppm               |
| Concentration of NO <sub>x</sub> (dry gas): | 300 ppm                | 300 ppm                |
| Gas flow (NTP):                             | 1.5 Nm <sup>3</sup> /h | 810 Nm <sup>3</sup> /h |
| Spray dryer inlet temperature:              | 150 °C                 | 150 °C                 |
| Adiabatic saturation temperature:           | 55 °C                  | 55 °C                  |
| Spray dryer outlet temperature:             | 64 and 74 °C           | Varying                |
| Gas residence time in spray dryer (NTP):    | 11 s                   | 16 s                   |
| Gas residence time in baghouse (NTP):       | 7 s                    | 9 s                    |
| Number of bags used:                        | 2                      | 20                     |
| Air-to-cloth ratio (NTP):                   | 0.017 m/s              | 0.020 m/s              |

TABLE 3. PROPERTIES OF THE SWEDISH  
ABSORBENTS UTILIZED

| Sorbent                        | Slaked lime                         | Limestone                          |
|--------------------------------|-------------------------------------|------------------------------------|
| Origin                         | Limhamn                             | Ignaberga                          |
| CaCO <sub>3</sub>              | 2 %                                 | 90 %                               |
| Ca(OH) <sub>2</sub>            | 90 %                                | -                                  |
| SiO <sub>2</sub>               | 2 %                                 | 7 %                                |
| Al <sub>2</sub> O <sub>3</sub> | 0.5 %                               | 0.5 %                              |
| Fe <sub>2</sub> O <sub>3</sub> | 0.3 %                               | 0.4 %                              |
| MgO                            | 1.0 %                               | 0.6 %                              |
| BET-surface area               | 14.4 m <sup>2</sup> g <sup>-1</sup> | 2.2 m <sup>2</sup> g <sup>-1</sup> |

## TECHNOLOGY DEVELOPMENT STATUS OF NITRIC OXIDE ABATEMENT USING FERROUS CHELATES

Donna J. Zabcik  
Dow Chemical U.S.A.  
Freeport, Texas

### ABSTRACT

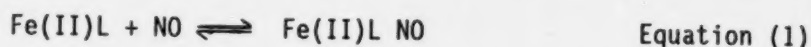
The absorption of nitric oxide from flue gas streams presents some unique problems due to its low solubility in conventional scrubbing solutions. The Dow patented process is based on the ferrous chelate chemistry investigated by the Japanese a decade ago. This chemistry is well documented to abate NO<sub>x</sub> to low levels; but commercialization was hindered due to high chemical makeup costs. The proposed Dow process has introduced several innovations which substantially reduce the operating costs to levels expected to be very competitive with commercially available technologies. The technical feasibility of this process to reach anticipated NO<sub>x</sub> emission requirements has been demonstrated both in laboratory studies and recently in field evaluations. A process description and the results of the field evaluations will be presented.

### INTRODUCTION

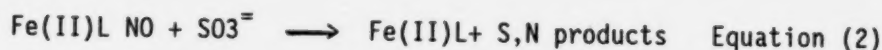
Techniques for NO<sub>x</sub> control fall into three broad categories: (1) fuel selection, (2) combustion modifications where NO<sub>x</sub> formation is minimized, and (3) flue gas treatment where NO<sub>x</sub> is actually removed from the gas stream. Until recently mechanical changes to the burner satisfied most emission requirements; however, air quality requirements in some parts of the country now require NO<sub>x</sub> removal from the flue gas streams. Dry methods such as thermal and catalytic reduction with ammonia have been demonstrated to achieve low NO<sub>x</sub> levels; however, a wet scrubbing technique is attractive since removal of both sulfur oxides and nitrogen oxides can be accomplished in a single process within existing wet FGD equipment. The process Dow has developed is based on liquid phase absorption of NO and electrochemical regeneration of the circulating solution. Laboratory and field demonstration results confirm that attention to proper scrubber design and chelate additive concentration allow flexibility to meet the most stringent NO<sub>x</sub> emission requirements.

## PROCESS

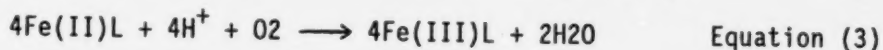
The wet scrubbing technique is based on the liquid phase reaction of ferrous iron with nitric oxide. Nitric oxide has a low solubility in aqueous solutions. The metal chelate enhances the absorption by reacting with the NO to form a nitrosyl complex which is soluble in the circulating solution (Equation 1).



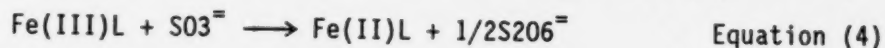
With certain chelating agents, the stability of the iron is increased since the ligand (identified as L in the equations) keeps the metal from precipitating. The complex then reacts with dissolved SO<sub>2</sub> to regenerate the metal chelate and to produce various nitrogen and sulfur compounds (Equation 2).



The concept of using metal chelates for NO<sub>x</sub> reduction is not a new one. The Japanese investigated it as a control method several years ago, but determined that high chemical makeup costs rendered the process uneconomical. More recently, a number of laboratories in the U.S. have pursued the development of this process because of the efficiency of the nitric oxide/iron chemistry. In the basic process an additive containing the metal chelate is introduced into the SO<sub>2</sub> scrubbing solution circuit. The nitrogen and sulfur compounds that are formed are removed from the process through a purge stream. Early research recognized the importance of the ionic state of the metal in the absorption step. Only the ferrous state of iron chelate is active with respect to NO<sub>x</sub> removal (1). Dissolved oxygen in the flue gas can oxidize the ferrous ions to ferric ions which are inactive for NO removal (Equation 3).



A competing reaction occurs as sulfite in the solution reduces the Fe(III) to Fe(II), but the reaction is very slow such that at equilibrium only about 10% of the total iron is in the active state (Equation 4).



In order to have enough Fe(II) to remove the NO<sub>x</sub> from the gas stream (Equation 1), a very high concentration of total iron was required in the circulating solution. Since the purge stream removing the sulfur and nitrogen compounds exits the circulating solution, that same high concentration of metal chelate was lost from the process. A major cost of operating the system was the chelate lost in the bleed from the scrubber circuit.

The patented Dow process is a method for reducing the iron chelate makeup requirements. The technique significantly reduces the total iron concentration by controlling the ionic state in the active Fe(II) form and unburdening the scrubber solution of the inactive Fe(III) load. The enhanced Fe(III) reduction method is accomplished using an electrochemical cell consisting of cathode and anode compartments separated by membranes (see Figure 1). When current is applied to the cell, the majority of the ferric chelate is reduced to ferrous chelate in the cathode and water is electrolyzed to oxygen and hydrogen ion at the anode. The process diagram in Figure 2 shows how the process could be integrated in a FGD plant.



Process costs have not been optimized. Capital requirements for NO abatement will be minimized for installations where a wet SO<sub>2</sub> scrubber exists. Operating costs of the process are strongly dependent on the flue gas conditions such as oxygen concentration, sulfur dioxide concentration, nitric oxide concentration, and overall abatement requirements. The process is particularly cost effective for coal and oil fired installations where the flue gas streams contain SO<sub>2</sub> and have low oxygen concentrations (2).

#### FIELD DEMONSTRATION

During the fall of 1987 a field trial was conducted to evaluate the technical viability of the Dow process. The objectives of the trial were to (1) maximize NO<sub>x</sub> abatement within a given scrubber system, (2) evaluate electrochemical cell performance under actual flue gas conditions, and (3) characterize the scrubbing solution purge stream. A worst case scenario field location was chosen to adequately demonstrate these parameters. In this evaluation a worst case scenario exists when the oxygen content of the flue gas stream is high and the flue gas does not contain sulfur. Those conditions maximize the conversion load on the electrochemical cell and increase chemical consumption rates. The conditions of the actual flue gas stream are listed below:

|                               |         |
|-------------------------------|---------|
| NO Concentration              | 40 ppmv |
| O <sub>2</sub> Concentration  | 14.5%   |
| CO <sub>2</sub> Concentration | 3.5%    |
| SO <sub>2</sub> Concentration | 0%      |
| Temperature                   | 170°F   |

Nitric oxide absorption was observed continuously as the NO concentration was spiked to as much as 700 ppmv. The field data confirmed literature reports that the absorption of NO is a mass transfer limited reaction (3). For this system increasing the Fe(II) concentration beyond 0.009 M had little effect on the abatement level. Increased abatement would require additional mass transfer stages.

Analyses showed that even under the severe conditions of high oxidation rates, the electrochemical cell maintained the majority of the iron chelate in the active Fe(II) state. Inspection of the electrodes and membranes at the end of the demonstration did not reveal significant wear or damage indicative of reduced lifetime.

A purge stream is taken from the scrubber solution to remove the sulfur and nitrogen compounds formed in the removal of SO<sub>2</sub> and NO. Characterization of this stream indicates the presence of one compound not found in a typical SO<sub>2</sub> scrubber purge. The additional reaction product is a nitrogen/sulfur salt identified as hydroxylamine disulfonate. The stream also contains iron chelate in the concentration circulated for NO removal.

## CONCLUSIONS

As NO<sub>x</sub> abatement requirements exceed the levels attainable by combustion modifications, a flue gas treatment technique must be employed. The benefits of a nitric oxide removal method that can be integrated into an existing FGD system are clear. The effectiveness of ferrous chelates to abate NO is well documented. Dow has demonstrated the successful use of an electrochemical cell to significantly reduce the operating costs of the process by controlling the ionic state of the iron chelate circulating solution. A number of parameters effect the efficient operation and design of the system. Additional field demonstrations will be used to determine an optimum system design.

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

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2. Tsai, S.S., Bedell, S.A., Kirby, L.H., Zabcik, D.J., "Field Evaluation of Nitric Oxide Abatement in Wet Flue Gas Using Ferrous Chelates", presented at the 1988 AIChE Summer Meeting, Denver, Colorado, 1988.
3. Tanaka, T., Koizumi, M., Yokoyama, T., Ishihara, Y., "Wet Process for Nitrogen Oxides Removal from Flue Gas (Part 4) - Study on the Reduction of EDTA-Fe(III) to EDTA-Fe(II) by Sodium Sulfite Solution," Denryoku Chuo Kenkyusho Hokoku, 1977.

FIGURE 1. Electrolytic Cell

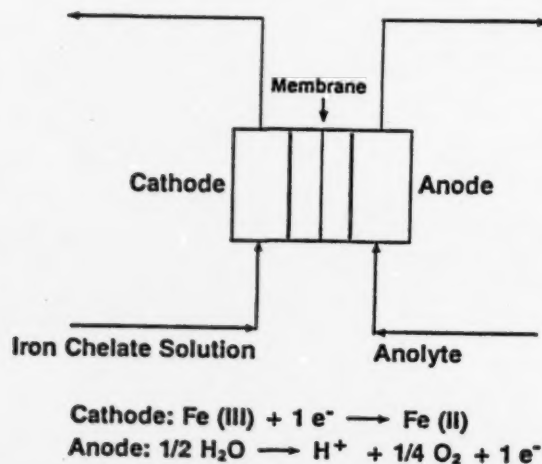
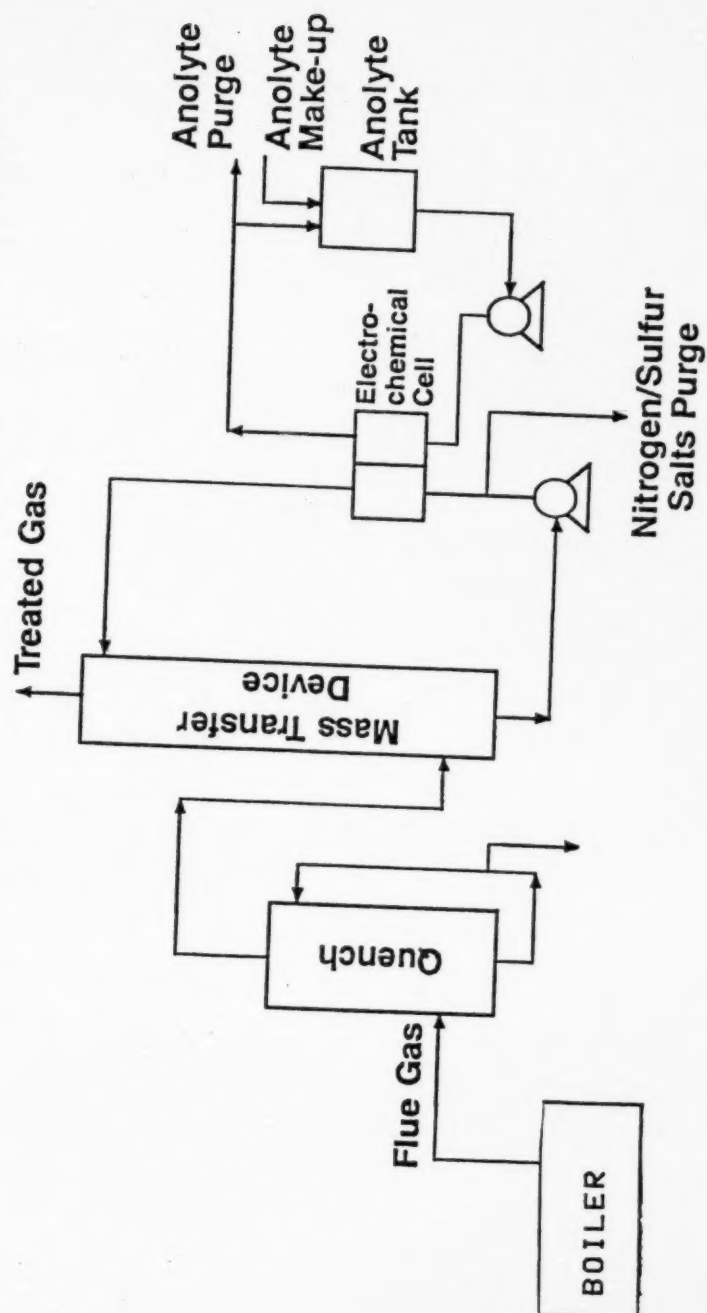


FIGURE 2

## Simplified Schematic of NOx Abatement Process



EXPERIENCES WITH ACTIVE COKE PROCESSES FOR  
SIMULTANEOUS REMOVAL OF SO<sub>2</sub> AND NO<sub>x</sub>

Ekkehard Richter, Karl Knoblauch, Wolfgang Wünnenberg  
Bergbau-Forschung GmbH  
Franz-Fischer-Weg 61  
4300 Essen 13  
Federal Republic of Germany

ABSTRACT

Active coke processes are used in the Federal Republic of Germany and in Japan for the simultaneous removal of SO<sub>2</sub> and NO<sub>x</sub>. They are based on adsorptive removal of SO<sub>2</sub> in a first stage and catalytic removal of NO<sub>x</sub> by ammonia in a second stage. The units are installed downstream of electrostatic precipitation and operated at temperatures between 100 and 180 °C so that no cooling and reheating of the flue gases is necessary. Loaded active coke is regenerated thermally, setting free SO<sub>2</sub> rich-gas which is processed to elemental sulfur or sulfuric acid.

Units are operated at a refinery and a lignite fired power plant since 1987. This paper describes the process and the operational experiences. Costs of the process are shown for a typical example.

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents does not necessarily reflect the views of the Agency and no official endorsement should be inferred.



## PROCESS DEVELOPMENT AND ACTIVE COKE QUALITY

Research and development of flue gas desulfurization processes using AC (active coke and activated carbon) started in the fifties. Laboratory and pilot tests showed high removal efficiencies. Also, the flue gases of coal and oil fired power plants were not cooled by the regenerative process installed downstream of electrostatic precipitation. No waste water resulted from the process. The  $\text{SO}_2$  rich-gas produced during thermal regeneration could be processed to chemical feeds like sulfuric acid or elemental sulfur. Nevertheless, the AC processes were applied industrially only in rare cases despite their advantages from an environmental point of view. Scrubbers and spray dryers dominated, producing gypsum or wastes to be deposited.

The breakthrough of the AC processes occurred when the BF-Process for simultaneous  $\text{SO}_2$  and  $\text{NO}_x$  removal was developed, based on a special active coke, Bevecar VA (Bergwerksverband GmbH, Essen, Fed. Rep. Germany). It is manufactured from bituminous coal. The diameter of the formed cylindrical pellets is 5 mm as a rule. Its special features are:

- o high adsorptive capacity for  $\text{SO}_2$ ,
- o fast adsorption kinetics,
- o high catalytic activity for  $\text{NO}_x$  removal by ammonia, even at low temperatures,
- o low reactivity towards oxygen,<sup>x</sup>
- o no activity losses due to regeneration,
- o high mechanical strength,
- o low price.

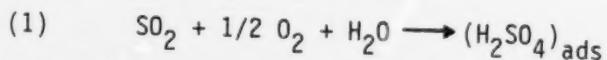
The last two properties result from the raw material, bituminous coal. High mechanical strength is necessary to restrict losses during transport in the process. The losses must be compensated by make-up active coke.

### MAIN CHEMICAL REACTIONS OF FLUE GAS CLEANING AND ACTIVE COKE REGENERATION

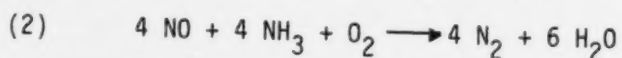
Adsorption and chemical reactions play an important role in the flue gas cleaning steps as well as the thermal regeneration of loaded active coke. A short summary is given below:

#### $\text{SO}_2$ ADSORPTION AND CATALYTIC $\text{NO}_x$ REDUCTION

Active coke adsorbs  $\text{SO}_2$  over a wide temperature range: from ambient to temperatures above 190 °C. The reaction of  $\text{SO}_2$  with oxygen and water vapor forming sulfuric acid (1) is catalyzed by carbon:

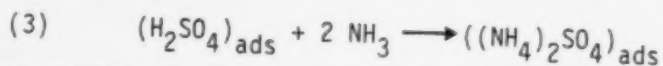


Maximum  $\text{SO}_2$  loads between 10 and 20 % by wt. are obtained as sulfuric acid has an extremely low vapor pressure. In the flue gases of coal and oil firings nitric oxides ( $\text{NO}_x$ ) consist mainly of nitrogen monoxide ( $\text{NO}$ ) and only of 5 to 10 % nitrogen dioxide ( $\text{NO}_2$ ). The latter is reduced quite fast by reaction with the carbon of the active coke at temperatures above 80 °C. When adding ammonia to the flue gases,  $\text{NO}$  is reduced catalytically on active coke:



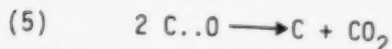
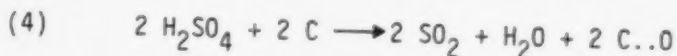
Any reactions between  $\text{NO}$  and  $\text{NH}_3$  to give ammonium nitrite will be excluded as it is unstable above 60 °C. It has been shown by long term experiments that even the formation of nitrate can be excluded at temperatures above 90 °C.

The concentrations were close to the detection limit (1). If the flue gases contain  $\text{SO}_2$  even after desulfurization sulfuric acid formed on the active coke is neutralized by ammonia:

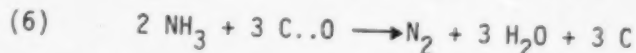


#### THERMAL REGENERATION OF ACTIVE COKE

The active coke loaded with sulfuric acid and corresponding ammonium salts needs thermal regeneration. From about 300 °C onwards sulfuric acid reacts with the carbon of the active coke to yield sulfur dioxide and carbon surface oxides C..O as an intermediate.



Ammonium sulfate decomposes by the reverse of reaction (3) to form sulfuric acid and ammonia. The latter will reduce  $\text{SO}_2$  spontaneously, an intermediate product of the  $\text{H}_2\text{SO}_4$  decomposition, or surface oxides:



Therefore, carbon losses decrease during thermal regeneration of active coke loaded with ammonium salts.

#### PROCESS FOR SIMULTANEOUS $\text{SO}_2$ AND $\text{NO}_x$ REMOVAL

This process is a further development of the original flue gas desulfurization process which has been applied in a demonstration plant at Steag-Kraftwerk Kellermann, Lünen, Fed. Rep. of Germany. This unit was operated at 130.000 m<sup>3</sup>/h (STP) flue gas.  $\text{SO}_2$  removal efficiencies of over 85 % were obtained in a one-stage moving bed adsorber. The regeneration of loaded active coke was carried out by mixing with hot sand. The  $\text{SO}_2$  rich-gas was processed to elemental sulfur in a modified Claus-plant or a Resox-plant (2). A second stage of  $\text{NO}_x$  removal was integrated into the process. This process will now be described.

#### FLUE GAS TREATMENT

The adsorber is installed downstream of the electrostatic precipitation (EP) and the blower. The flue gas temperature is usually between 120 and 150 °C. The dust content is under 150 mg/m<sup>3</sup> (STP). Flue gas from precipitation is cooled by water injection in a gas cooler to a temperature of about 120 °C. A two-stage moving bed adsorber is used for removal of  $\text{SO}_2$  and  $\text{NO}_x$  as shown in Fig. 1. Additionally, halogen compounds - like HCl and HF -, arsenic, selenium, and mercury are removed by adsorption on active coke. The concept of the adsorber will be described in more detail later on. The cleaned flue gas leaves the adsorber and streams to the stack. Active coke in the adsorber is loaded with sulfate and regenerated in the desorber.  $\text{SO}_2$  rich-gas is produced in the latter. The rich-gas is cleaned and processed to sulfuric acid in a separate unit. The units - adsorber, desorber, rich-gas cleaning, and sulfuric acid plant - are described in more detail.

The adsorber consists of two stages. There is a cross flow of active coke (from the top to the bottom) and flue gas (horizontally) through the moving beds. Ammonia is injected into the flue gases between the stages. Flue gas streams through the first (lower) bed for  $\text{SO}_2$  removal and then through the second (upper) bed for the removal of  $\text{NO}_x$  and residual  $\text{SO}_2$ . Regenerated active coke is introduced into

the adsorber unit at the top of the second stage. At the bottom of the second stage it is loaded with a small amount of ammonium sulfate. In the first stage it is loaded additionally with sulfuric acid formed in the  $\text{SO}_2$  adsorption step. The loaded active coke is drawn off from the bottom of the first stage and conveyed to the thermal regeneration in the desorber.

The construction of the adsorber is shown in Fig. 2. The flue gas duct enters the center of two units facing each other. The gas streams from the inner chamber through the beds to two separate outer chambers. There it is collected and flows through a vertical duct where ammonia is injected. The two flue gas streams enter the outer chambers of the second stage, stream through the beds, flow together in the inner chamber and to the stack. The active volume of each adsorber bed is flown through by the flue gas. The gas inlets and outlets are formed by louvers. Additionally, conical ducts at the top and the bottom of the bed serve to supply and draw-off active coke. Three segments are arranged in parallel. Each segment has three cones at the top and the bottom. The active coke is discharged at the bottom onto a conveyor belt (3).

#### ACTIVE COKE REGENERATION

The regeneration of loaded active coke is carried out in the desorber. Several types can be used. Tube-type desorbers are used in the large scale plants. Active coke moves through tube bundles outside-heated by combustion gases from a burner unit. Part of the cooled combustion gas is recycled to decrease the inlet temperature. Active coke is heated to temperatures of about  $400^\circ\text{C}$ . After heating up the active coke it reaches a degassing chamber where the  $\text{SO}_2$  rich-gas leaves the desorber. The rich-gas contains 25 - 30 % by vol. of  $\text{SO}_2$ ,  $\text{CO}_2$ , water vapor,  $\text{HCl}$ ,  $\text{HF}$ , and trace elements. The processing of the gas is described later on. Subsequently, the active coke is cooled in a second tube bundle to temperatures of about  $120^\circ\text{C}$ . Dust and particles below a certain sizes are sieved off. Active coke losses due to chemical losses and abrasion are compensated by make-up AC before the active coke is conveyed to the hopper above the adsorber.

#### PURIFICATION OF $\text{SO}_2$ RICH-GAS

Treatment of the  $\text{SO}_2$  rich-gas includes purification and subsequent processing to elemental sulfur or sulfuric acid. Besides  $\text{SO}_2$ , rich-gas leaving the desorber contains about 50 % water vapor, 20 %  $\text{CO}_2$  and less than 1 %  $\text{NH}_3$  (all by volume). In case of treating combustion gases of coal minor concentrations of halogenes, selenium, arsenic, mercury, and dust are also present. The flow scheme of the rich-gas purification is shown in Fig. 3. The gas is led through two scrubbers to remove  $\text{As}_2\text{O}_3$ , Se, Hg, and ammonia by reaction with sulfuric acid (4). The halogens are dissolved in the condensed water from the subsequent gas cooler. The condensed water is either neutralized by sodium hydroxide and fed into the flue gas quencher or it is vaporized e.g. crystallized. Sulfuric acid aerosols are removed by electrostatic precipitators. The last unit is a safety filter containing activated carbon to removal possible traces of mercury. The solutions of the two scrubbers are circulated. Solid products are removed from the solution by a filter. The filter cake has to be disposed in a special disposal area. In this way it is possible to remove these substances completely without producing waste water. (Wet FGD processes may produce large amounts of waste water.)

#### PROCESSING OF $\text{SO}_2$ RICH-GAS

The  $\text{SO}_2$  rich-gas may subsequently be processed to elemental sulfur, sulfuric acid or liquid sulfur dioxide. Simplified flow schemes of the processes are shown in Fig. 4. For sulfur production, part of the  $\text{SO}_2$  is reduced to  $\text{H}_2\text{S}$  and sulfur in



a reduction burner.  $\text{H}_2\text{S}$  and  $\text{SO}_2$  are converted in a Claus unit. Liquid sulfur is drawn off from the condensers. Concentrated sulfuric acid is produced by catalytic oxidation after the addition of air. The production of liquid sulfur dioxide is carried out by means of coolers, dryers and a  $\text{CO}_2$  stripping column.

If the process for simultaneous  $\text{SO}_2$  and  $\text{NO}_x$  removal is installed at a site where a sulfuric acid or a Claus-plant is already operated the  $\text{SO}_2$  rich-gas can be fed into this unit after gas purification. As a rule the size of these plants is sufficient to take up the total  $\text{SO}_2$  amount from the flue gas treatment plant without modifications.

### PROCESS APPLICATIONS

The lay-out data for plants operated already or under construction are shown on Tab. 1. In every case,  $\text{SO}_2$  is removed almost completely. The design of the  $\text{NO}_x$ -stage is adapted to the necessities of  $\text{NO}_x$ -emission control. Uhde GmbH, Dortmund, and Mitsui Mining Company Ltd., Tokyo, as well as Kinetics Technology International B.V., Zoetermeer (NL) and Steag AG, Essen are licencees of Bergbau-Forschung GmbH, Essen.

At Aichi-Refinery, the plant cleans the flue gases of a fluidized catalytic cracker unit. The plant is operated since May 1987, only interrupted by usual revision. The removal efficiencies are higher than predicted. The residual  $\text{NH}_3$ -content in the off-gases is under  $20 \text{ mg/m}^3$ . A photograph of the plant is shown on Fig. 5. The  $\text{NO}_x$ -stage is installed at the top of the  $\text{SO}_2$ -stage. The desorber unit is placed in front of the two stages. Active coke is conveyed by conveyer belts at the bottom of the adsorber sections and by bucket elevators from the bottom of the adsorber to the top of the desorber resp. the bottom of the desorber to the top of the adsorber. The removal efficiencies for  $\text{SO}_2$  and  $\text{NO}_x$  are higher as the design values. The active coke losses are under the guaranteed values.

The lignite fired boilers 5 and 7 at EVO Kraftwerk Arzberg are located in a distance of about 150 m. The adsorbers are installed directly at the boilers as shown on Fig. 6. They contain two stages, one above the other, as shown on Fig. 2. One central desorption with two parallel units and the respective  $\text{SO}_2$ -rich-gas cleaning and sulfuric acid units are located nearby the desorption (3).

The active coke transport from adsorption to desorption and back again occurs by low velocity pneumatic conveyers. This method was chosen because of the long distance between the adsorber for boiler 7 and the central desorber. Cleaned flue gas is compressed and used for the transport to hoppers at the top of the adsorbers resp. desorbers. About  $24 \text{ m}^3/\text{h}$  of active coke are conveyed by each line.

The first start-up of adsorber unit 7 was in July, 1987, of unit 5 in Sept., 1987. Periods of operation and of interrupt for maintainance and improvement alternated during the subsequent year. Excellent removal efficiencies were obtained during normal operation. Usually, the outlet concentration of  $\text{SO}_2$  were under  $100 \text{ mg SO}_2/\text{m}^3$  (STP), those for  $\text{NO}_x$  under  $200 \text{ mg NO}_x/\text{m}^3$  (STP) complying the German regulations. Until May 1988, 1400 t of high purity 98 % sulfuric acid was produced and sold as chemical feed.

The problems of the plant arose mainly by the pneumatic conveying of active coke. First of all, the conveying system worked unreliably. Some time was necessary to optimize pulse pressures and times for the tubes of different length, delivery head and capacity. Additionally, abrasion as well as wear were too high. Within a limited time a lot of tubes, especially at right angle bends and switches were wearied out. It was necessary to line all these tubes at the inner wall by molten basalt. By this method the walls were protected but the abrasion of active coke increased due to the initial surface roughness.



Much more dust and fine particles than expected were produced by the pneumatic conveying system and its receivers. The measures for wind sieving of the fines were not sufficient to protect the adsorber against deposition of fines in areas of poor particle motion. An increased pressure drop was observed due to increased flow resistance and non-uniform flow distribution over the adsorber cross section.

The operation of the plant was interrupted in May, 1988 for completion resp. improvements of the following units: Pneumatic conveyer, wind sieving, sieving of fines, and active coke distribution from the receiver of the pneumatic conveyer to the adsorber segments. The plant starts its operation again at the end of October, 1988.

As shown on Tab. 1, a plant for  $\text{SO}_2$  and  $\text{NO}_x$ -removal by BF/Uhde/Mitsui-Process is under construction at Hoechst AG, Frankfurt (West-Germany) in the meantime.

Sumitomo Heavy Industries, Tokyo has developed a process for simultaneous  $\text{SO}_2$ - and  $\text{NO}_x$ -removal which is similar to the BF-Process.  $\text{SO}_2$ - and  $\text{NO}_x$ -removal occurs in one stage upstream of which ammonia is injected. The  $\text{SO}_2$ -removal efficiency is increased by this measure, but the  $\text{NO}_x$ -removal efficiency is limited to about 50 % or less depending on the  $\text{SO}_2$  and  $\text{NO}_x$  concentrations and the gas temperature. After operation of a demonstration plant for 300 000  $\text{m}^3/\text{h}$  (STP) flue gas at Matsushima, a commercial plant for  $\text{SO}_2$  removal was constructed at Nippon Steel's Nagoya Works. The unit removes > 95 % of  $\text{SO}_2$  from 900 000  $\text{m}^3/\text{h}$  (STP). The  $\text{SO}_2$  rich-gas is processed to sulfuric acid in an existing sulfuric acid plant. A simplified scheme is shown on Fig. 7. Active coke, manufactured by Bergwerksverband, Essen, is used. The operation follows the design. The active coke losses are very low as AC transport occurs by conveyer belts and bucket elevators.

#### COSTS OF THE UHDE/BF/MITSUI-PROCESS FOR SIMULTANEOUS $\text{SO}_2/\text{NO}_x$ -REMOVAL

In the past, several cost studies have been presented showing the advantages of the different processes for  $\text{SO}_2$ - and  $\text{NO}_x$ -removal. Experience shows that cost calculations may serve as examples only. Which processes cost less than others often depends on special assumptions which prevent a serious comparison.

In this section, the costs of the process for simultaneous  $\text{SO}_2$ - and  $\text{NO}_x$ -removal are summarized. The calculations were carried out by Uhde GmbH (4). Based on German price conditions in 1988, the specific investment costs amount to approx.

175 - 225 US \$/kW<sub>e1</sub> (1 US \$ = 1.85 Deutsche Mark)

|                                         |                                                                                            |
|-----------------------------------------|--------------------------------------------------------------------------------------------|
| Capacity of power plant:                | 300 MW <sub>e1</sub>                                                                       |
| Type of fuel:                           | Bituminous coal                                                                            |
| S-content of the fuel:                  | 1.3 % by weight                                                                            |
| Type of boiler:                         | Dry bottom boiler                                                                          |
| $\text{NO}_x$ -conc. of the flue gases: | 900 mg $\text{NO}_2/\text{m}^3$ (STP), dry basis<br>based on 6 % by vol. $\text{O}_2$      |
| Emissions standards $\text{SO}_2$ :     | less 200 mg $\text{SO}_2/\text{m}^3$ (STP), dry basis<br>based on 6 % by vol. $\text{O}_2$ |
| $\text{NO}_x$ :                         | less 200 mg $\text{NO}_2/\text{m}^3$ (STP), dry basis<br>based on 6 % by vol. $\text{O}_2$ |
| Flue gas inlet temperature:             | less than 160 °C                                                                           |
| End product:                            | sulfuric acid.                                                                             |

The above-mentioned investment costs include the following supplies and services for a flue gas treatment plant:

- basic and detail engineering
- licence fees
- process equipment, electrical equipment and instrumentation

- civil works
- erection
- first filling with active coke
- commissioning.

Not included in the above mentioned estimate are the costs for:

- real estate
- site development including surface reinforcement and foundations
- capital expenditure before start-up
- engineering of the costumer
- costs for obtaining official permits.

Fig. 8 shows the breakdown of the investment costs. 65 % of the total investment costs are necessary for SO<sub>2</sub> removal consisting of following process sections.

- adsorber (SO<sub>2</sub>-stage)
- desorber
- active coke transport system
- processing of SO<sub>2</sub> rich-gas
  - rich-gas purification
  - sulphuric acid production
- storage systems for active coke and sulphuric acid

Only 35 % of total investment costs have to be taken into consideration for the additional NO<sub>x</sub>-reduction consisting of the following process sections:

- adsorber (NO<sub>x</sub>-stage)
- ammonia storage and supply
- additional active coke transport system

In the most cases, however, the determining factor for the choice of a process is the total operating costs. Based on the above design criteria and investment costs, the operating costs for a plant with 5000 operating hours per year (full load) amount to approx.

10.8 mills

This cost calculation is based on following specific costs:

- capital service incl. insurance and taxes:  
13.5 % of the investment costs
- maintenance: 3 % of the investment costs  
including the first filling with active coke
- personnel: 16 employees at 40.000 US \$/year
- active coke: 1,080 US \$/t
- electric power: 0.069 US \$/kWh
- ammonia: 0.24 US \$/kg
- fuel (44,300 kJ/kg): 0.24 US \$/kg
- sulphuric acid (96 %): 0.035 US \$/kg

Fig. 9 shows the breakdown of the operating costs. 63 % of the operating costs are fixed expenses because capital service, maintenance and personnel are independent of the operating hours of the plant. The remaining of 37 % are variable, being related to the operating time of the plant.

Furthermore it can be seen that only approx. 27 % of the operating costs are necessary for the reduction of NO<sub>x</sub>. The above-mentioned operating costs of 10.8 mills

can therefore be split up into 2.7 mills for the reduction of  $\text{NO}_x$  and 8.1 mills for the removal of  $\text{SO}_2$ .

Based on our market experience, the operating costs for the simultaneous process are comparable or even lower than the costs of competitive processes, e.g. wet limebased  $\text{SO}_2$  scrubbing with gipsum production combined with a SCR process for  $\text{NO}_x$ -reduction. Compared to the other processes, the  $\text{SO}_2$  removal part of the Uhde/BF/Mitsui-Process can be more expensive while the  $\text{NO}_x$ -reduction part is less costly.

#### CONCLUSIONS

The Uhde/BF/Mitsui-Process for simultaneous  $\text{SO}_2$ - and  $\text{NO}_x$ -removal is one of the most advanced processes for flue gas cleaning, removing not only  $\text{SO}_2$  and  $\text{NO}_x$  but also  $\text{HCl}$ ,  $\text{HF}$ , selenium and arsenic compounds, mercury and dust from the flue gases. The main advantages of the process are:

- High separation efficiencies for  $\text{SO}_2$  and  $\text{NO}_x$
- Usable end products: sulphur,  $\text{H}_2\text{SO}_4$  or  $\text{SO}_2$  (liquid)  
hence, no dumping problems
- No waste water, hence, problems are not transferred from the flue gas into the waste water

These advantages can be achieved with operating costs which are comparable or even lower than those for competitive processes.

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4. Richter, E.; Henkel, J.: The dry BF-process for simultaneous  $\text{SO}_2/\text{NO}_x$ -removal. Executive Body for the Convention of Long-range Transboundary Air Pollution: Workshop on Emission Control Costs, Esslingen, Sept. 28 - Oct. 1, 1987., pp. 201/217



| Supplier<br>Plant                             | Uhde GmbH<br>Kraftwerk Arzberg<br>Lignite Fired Boiler<br>Boiler 5      Boiler 7 |         | Mitsui Mining Comp.<br>Alchi Raffinerie<br>FCC Off-gases |  | Uhde GmbH<br>Hoechst AG<br>Trivalent Boiler<br>Hard Coal      Gas |         |
|-----------------------------------------------|----------------------------------------------------------------------------------|---------|----------------------------------------------------------|--|-------------------------------------------------------------------|---------|
| Flue Gas Volume<br>(m <sup>3</sup> /h (I.N.)) | 451 000                                                                          | 658 000 | 236 000                                                  |  | 323 000                                                           | 278 000 |
| Temperature<br>(°C)                           | 150                                                                              | 150     | 190                                                      |  | 140                                                               | 120     |
| Inlet:                                        |                                                                                  |         |                                                          |  |                                                                   |         |
| SO <sub>2</sub> (mg/m <sup>3</sup> , dry)     | 3 800                                                                            | 3 600   | 750                                                      |  | 2 500                                                             | 2       |
| SO <sub>3</sub> (mg/m <sup>3</sup> , dry)     | 125                                                                              | 125     | 110                                                      |  | 210                                                               | -       |
| NO <sub>2</sub> (mg/m <sup>3</sup> , dry)     | 750                                                                              | 750     | 205                                                      |  | 900                                                               | 450     |
| dust (mg/m <sup>3</sup> )                     | 400                                                                              | 150     | 60                                                       |  | 85                                                                | -       |
| Removal Efficiencies:                         |                                                                                  |         |                                                          |  |                                                                   |         |
| SO <sub>x</sub> (%)                           | >95                                                                              | >95     | >90                                                      |  | >90                                                               | -       |
| NO <sub>x</sub> (%)                           | >73                                                                              | >73     | >60                                                      |  | >78                                                               | >75     |
| Residual Contents:                            |                                                                                  |         |                                                          |  |                                                                   |         |
| Dust (mg/m <sup>3</sup> )                     | 50                                                                               | 50      | 20                                                       |  | 50                                                                | -       |
| NH <sub>3</sub> (mg/m <sup>3</sup> , dry)     | <35                                                                              | <35     | <35                                                      |  | <35                                                               | <35     |
| Start-up:                                     | 7/1987                                                                           | 9/1987  | 5/1987                                                   |  |                                                                   | 1990    |

Table 1: BF-Processes for SO<sub>2</sub>- and NO<sub>x</sub>-Removal from Flue Gases-Plants under Construction and in Operation

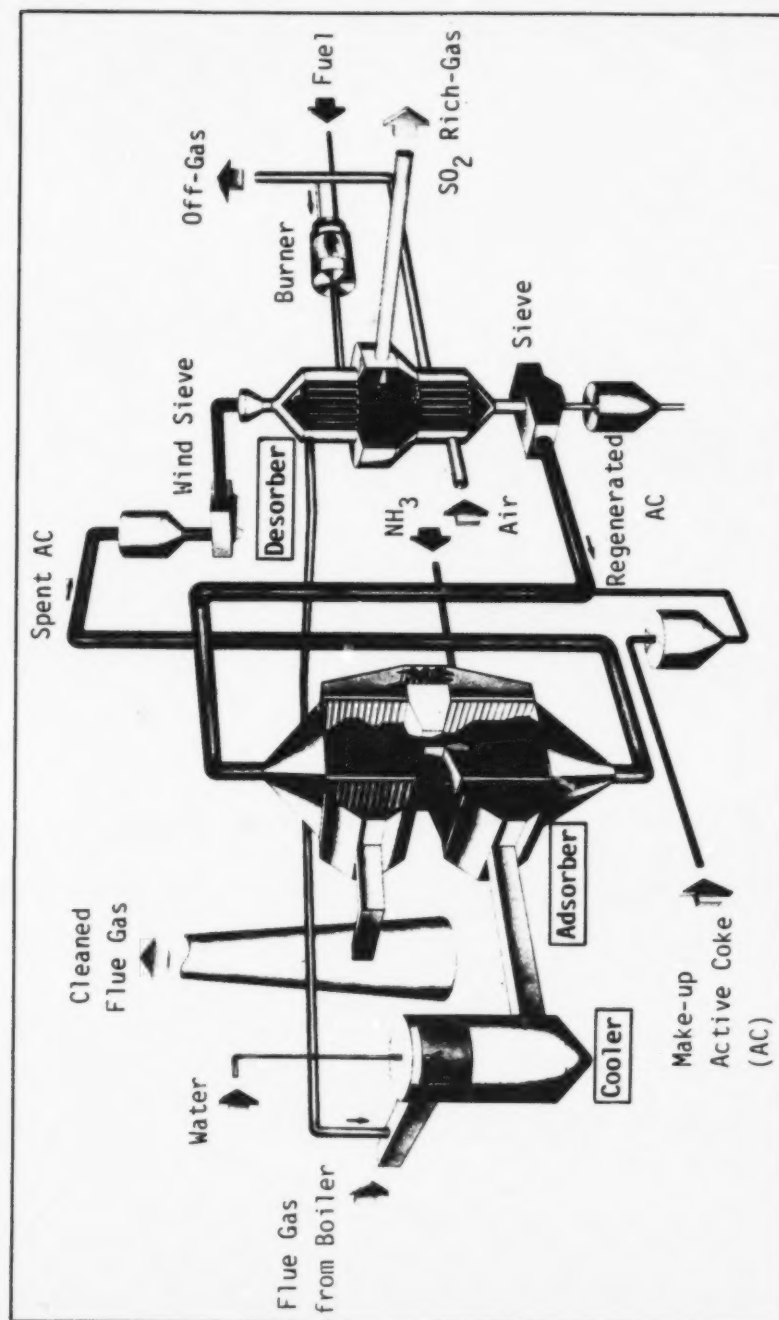


Figure 1: Simplified Scheme of the Uhde/BF/Mitsui-Process for  $\text{SO}_2$ - and  $\text{NO}_x$ -Removal

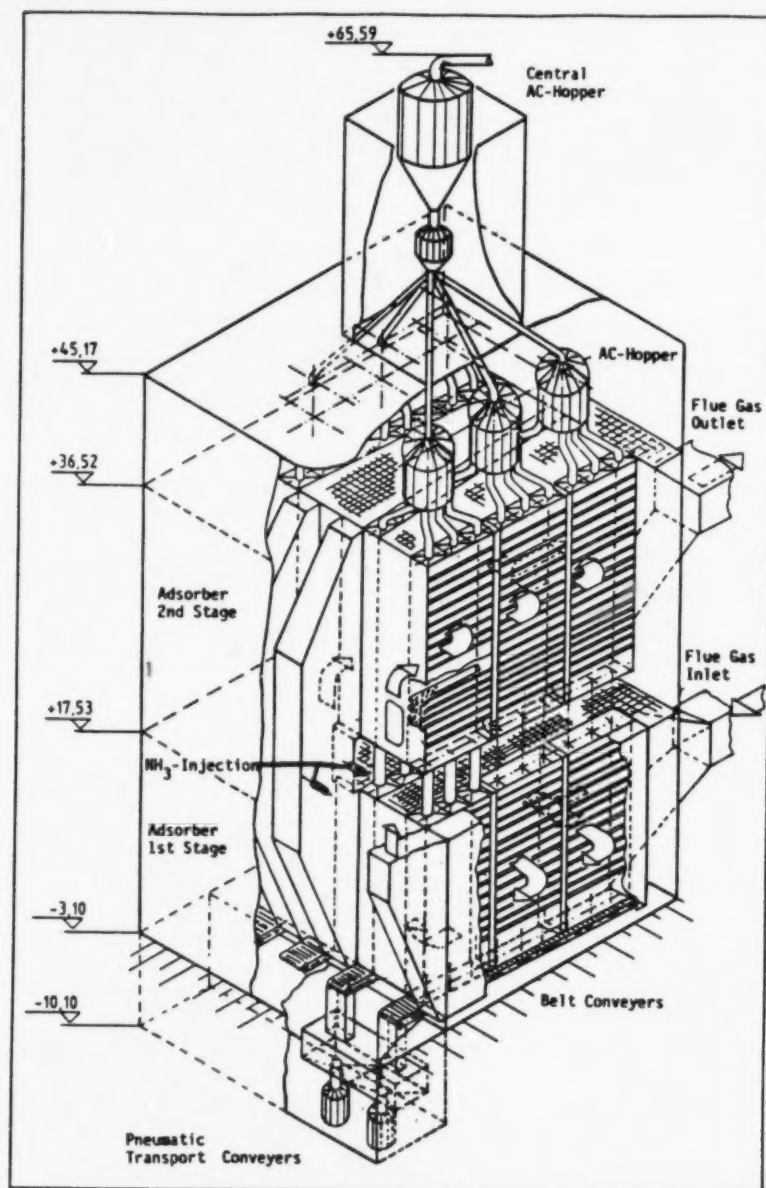


Figure 2: Perspective View of the Two-stage Adsorber for  $\text{SO}_2$ - and  $\text{NO}_x$ -Removal

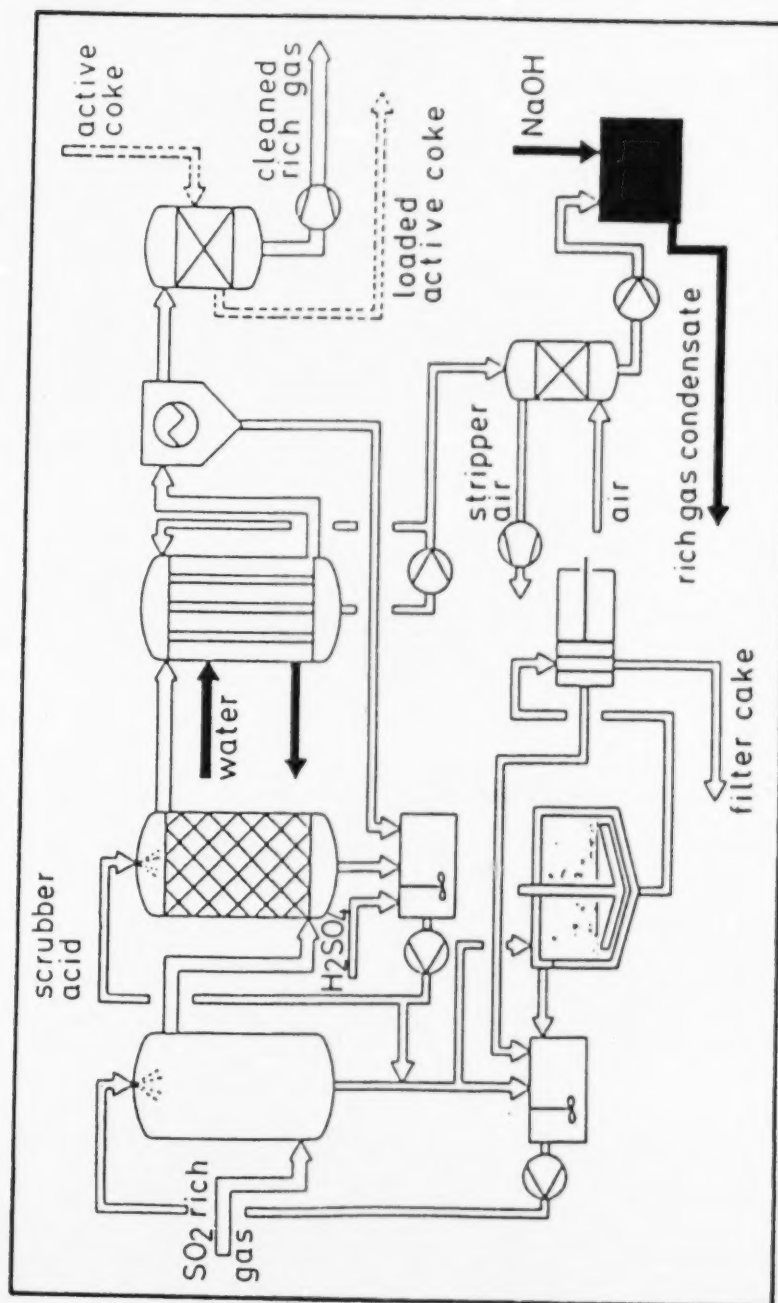
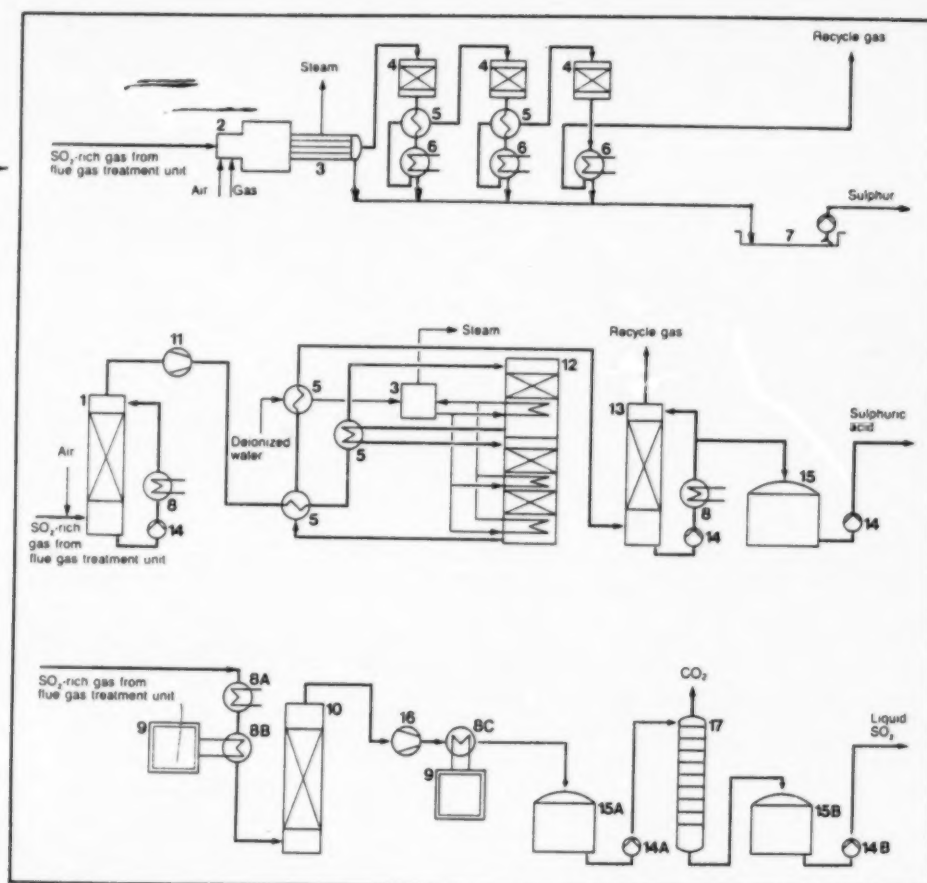


Figure 3: Scheme of SO<sub>2</sub> Rich-Gas Purification before Processing to Sulfuric Acid





- |                              |                                   |                             |
|------------------------------|-----------------------------------|-----------------------------|
| 1 Drying tower               | 7 Sulphur pit                     | 14 Pump                     |
| 2 Reduction burner           | 8 Cooler                          | 15 Storage tank             |
| 3 Waste heat boiler          | 9 Refrigeration set               | 16 Compressor               |
| 4 Claus 3-stage catalyst bed | 10 Drier                          | 17 CO <sub>2</sub> stripper |
| 5 Heat exchanger             | 11 Blower                         |                             |
| 6 Condenser                  | 12 SO <sub>2</sub> catalytic unit |                             |
|                              | 13 Adsorber                       |                             |

Figure 4: Processes for the Production of Elemental Sulphur, Sulphuric Acid and Liquid SO<sub>2</sub> from SO<sub>2</sub> Rich-Gas

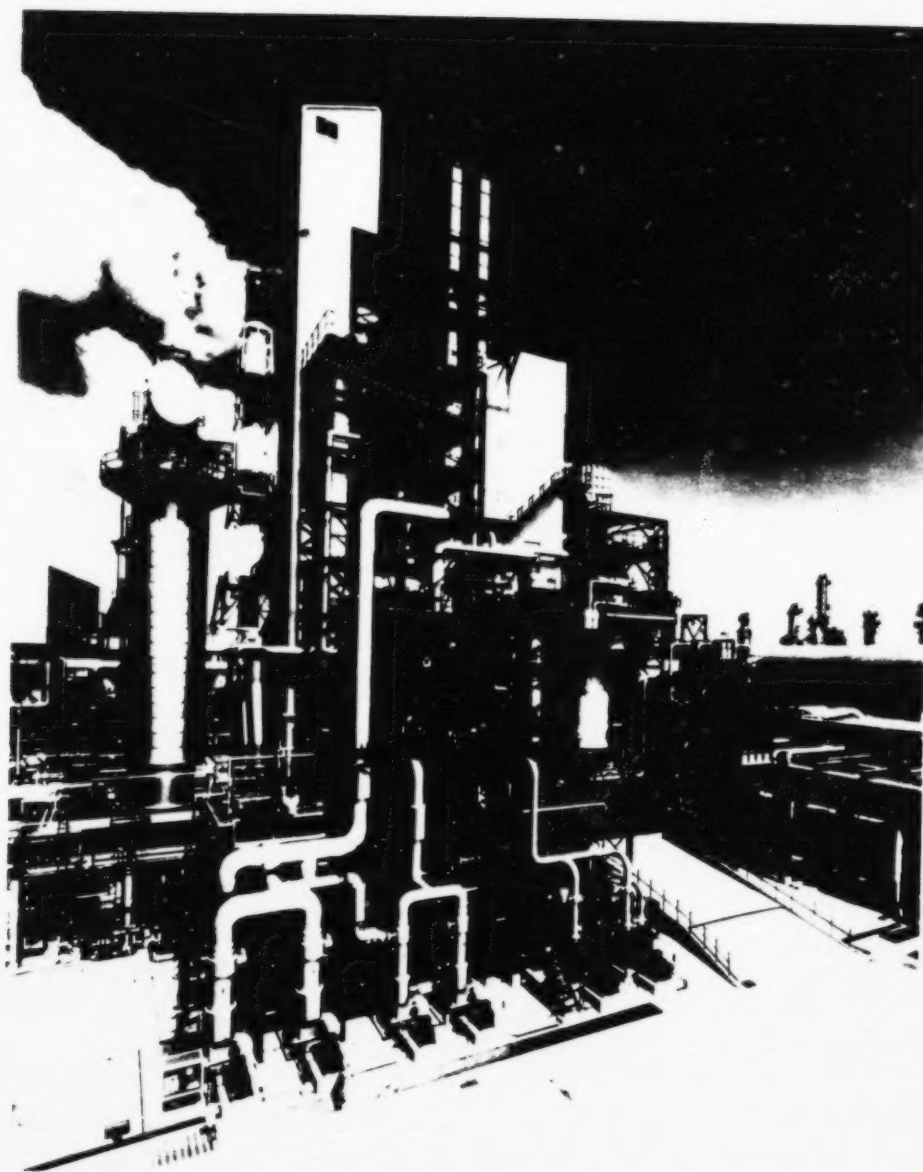


Figure 5: Plant for  $\text{SO}_2$ - and  $\text{NO}_x$ -Removal by BF/Mitsui-Process at Idemitsu's Aichi Refinery, Nagoya (Japan)

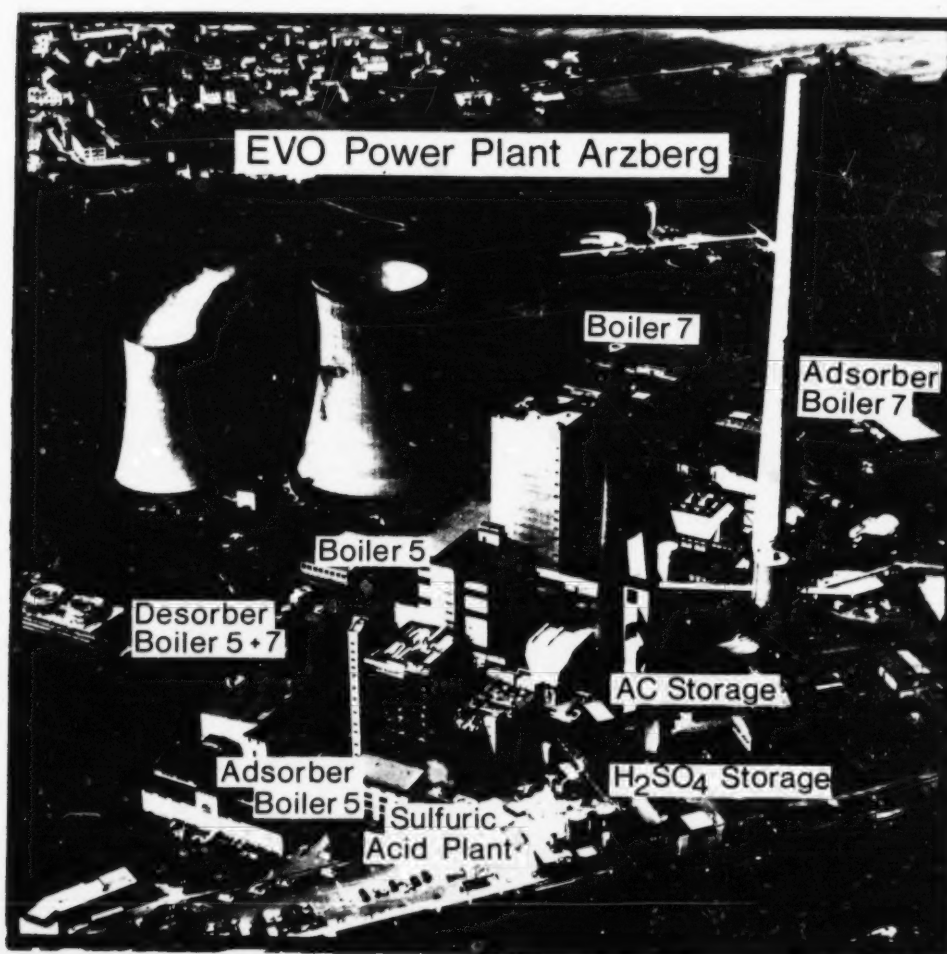


Figure 6: Aerial Photograph of EVO's Arzberg Power Plant and  $\text{SO}_2/\text{NO}_x$ -Removal Units by BF/Uhde/Mitsui

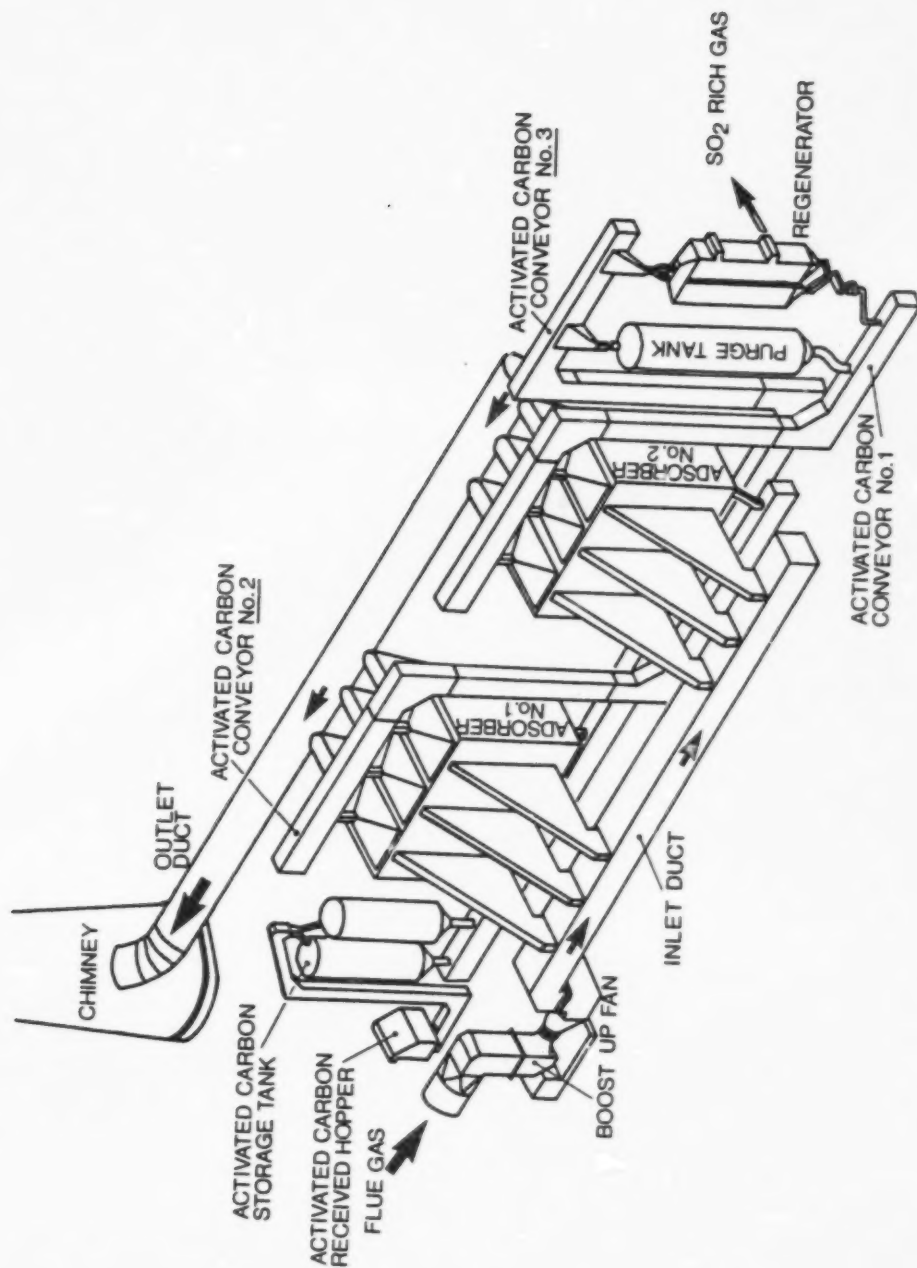


Figure 7: Simplified Scheme of the Flue Gas Treatment Plant Constructed by Sumitomo Heavy Industries at Nippon Steel's Nagoya Works



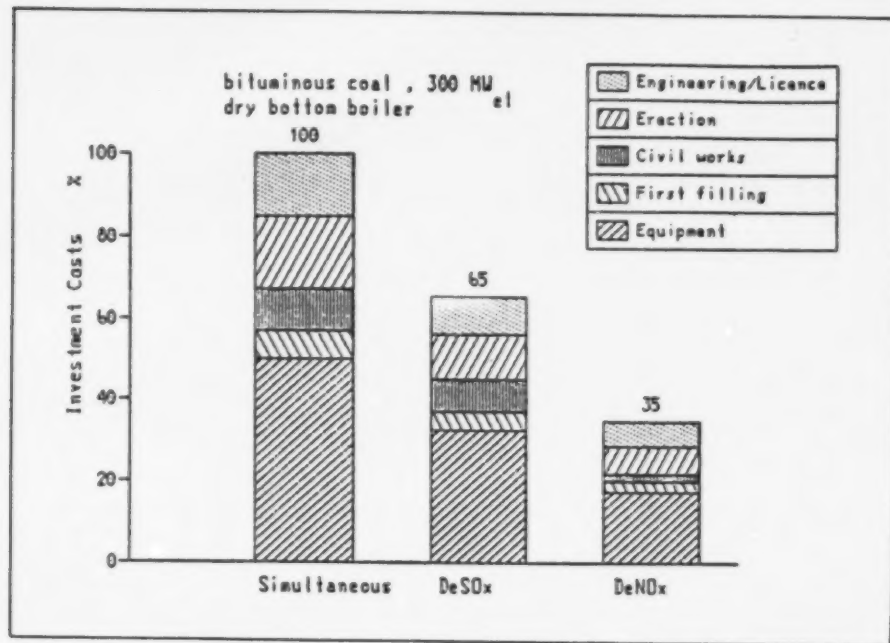


Figure 8: Breakdown of the investment costs

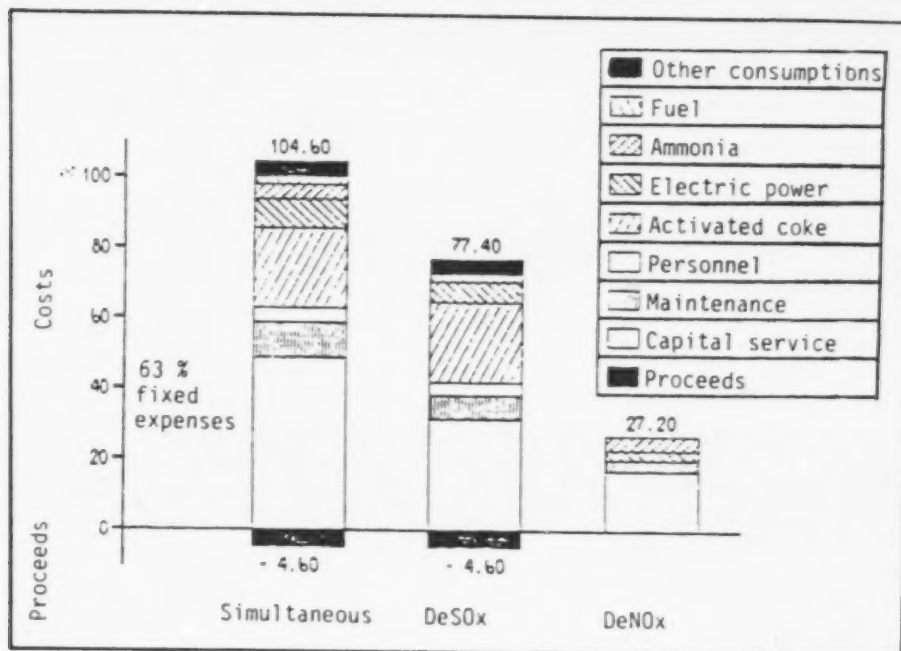


Figure 9: Breakdown of operating costs

## SIMULTANEOUS NO<sub>x</sub>/SO<sub>2</sub> REMOVAL WITH DRY SORBENT INJECTION\*

M. E. Teague  
R. E. Thompson  
L. J. Muzio  
Fossil Energy Research Corp.  
23342 C South Pointe  
Laguna Hills, CA 92653

### ABSTRACT

Dry SO<sub>2</sub> removal processes using calcium hydroxide have yielded nominally 40-60 percent SO<sub>2</sub> removal at Ca/S ratios of 2. Efforts to increase SO<sub>2</sub> removal and calcium utilization by modifying hydrate properties have met with only marginal success. However, instead of directly increasing SO<sub>2</sub> removal and calcium utilization, the attractiveness of dry SO<sub>2</sub> removal processes may also be enhanced if NO<sub>x</sub> could be removed along with SO<sub>2</sub> using a single dry sorbent. To increase the cost-effectiveness of this process, a modified hydrate was developed that simultaneously removes SO<sub>2</sub> and NO<sub>x</sub>. Pilot-scale results of injecting this single dry powder into a furnace are presented to characterize the SO<sub>2</sub> and NO<sub>x</sub> removal characteristics, and a preliminary economic analysis of this process is developed. Initial pilot-scale results indicate that the dry sorbent is capable of simultaneous SO<sub>2</sub> and NO<sub>x</sub> removals of 50 percent at Ca/S=2 and urea/NO<sub>x</sub>=0.5. Higher SO<sub>2</sub> and NO<sub>x</sub> removals can be achieved by changing the Ca/S and/or urea/NO<sub>x</sub> ratios. The simplicity of the process is an important factor as it potentially offers an inexpensive approach for retrofit applications where both SO<sub>2</sub> and NO<sub>x</sub> can be removed with minimum capital expenditures.

### DISCLAIMER

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

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## INTRODUCTION

### BACKGROUND

A major environmental problem confronting industry and the utilities is acid rain and the emissions of  $\text{SO}_2$  and  $\text{NO}_x$ , particularly from coal-fired plants in the Midwest and Northeastern United States. Since many of the plants were placed in service prior to the implementation of New Source Performance Standards (NSPS) for emissions control, any large scale reduction in  $\text{NO}_x$  and  $\text{SO}_2$  emissions will require the application of control equipment to these older plants on a retrofit basis. The projected impact to the U.S. utility industry (and consumers) of applying conventional  $\text{SO}_2$  control equipment, such as wet scrubbers, is measured in billions of dollars. While the initial focus on acid rain was  $\text{SO}_2$  emissions, recent research has also shown that nitrogen oxide ( $\text{NO}_x$ ) emissions may be a significant factor in forest damage via a different mechanism than  $\text{SO}_2$ . As a direct result, Germany has passed some of the most stringent  $\text{NO}_x$  regulations for existing plants in the world. Therefore in the future, utilities may face the prospect of not only a large expensive retrofit program for  $\text{SO}_2$  control but also the potential additional expense of  $\text{NO}_x$  control.

Retrofit  $\text{SO}_2$  control technology has traditionally focused on wet and spray dry scrubbing processes to reduce the  $\text{SO}_2$  emissions. These processes, however, require a large capital investment and complex hardware. They also require adequate space at the existing facility to install the scrubber hardware. Dry sorbent injection control technologies simplify the  $\text{SO}_2$  removal process by eliminating the wet slurry, liquid waste treatment systems, and extensive additional hardware of wet scrubbing processes, resulting in lower capital costs (1, 2). From a cost basis, the lower capital cost (albeit higher operating cost) of dry sorbent injection systems are potentially attractive for older units with a limited, or uncertain, remaining life.

Just as there are a spectrum of approaches to limit  $\text{SO}_2$  emissions from stationary sources, a similar range of approaches exist for  $\text{NO}_x$  control. From the standpoint of  $\text{NO}_x$  control alone, combustion modifications (i.e. low- $\text{NO}_x$  burners, staged combustion via overfire air, or reburning) generally afford the least capital intensive approach, with  $\text{NO}_x$  reductions of the order of 30-50 percent (3). At the other end of the spectrum, selective catalytic  $\text{NO}_x$  reduction with ammonia offers the potential for the highest  $\text{NO}_x$  removal--potentially as high as 90 percent--but also is capital intensive (3). Intermediate between combustion modifications and selective catalytic  $\text{NO}_x$  reduction are selective non-catalytic  $\text{NO}_x$  reduction processes. These processes react  $\text{NO}_x$  with a compound such as  $\text{NH}_3$  (Thermal De $\text{NO}_x$  (4)), urea (5, 6), or cyanuric acid (7) in a temperature region of 800-1100°C.

With the current motivation to simultaneously reduce both  $\text{NO}_x$  and  $\text{SO}_2$  emissions, a dry sorbent injection process that can remove  $\text{SO}_2$  and  $\text{NO}_x$  with a single cost effective sorbent is an attractive option for retrofit applications. A process based on injecting a lime-urea hydrate into the combustion gases in the upper furnace region of a boiler was recently developed by Fossil Energy Research Corp. (8). The lime-urea hydrate is prepared by dissolving a specified amount of urea in water creating a solution that is used to hydrate a quicklime. Figure 1 shows a schematic of the process.

This hydration process produces a fine, dry powder sorbent containing calcium hydroxide and urea capable of removing both  $\text{NO}_x$  and  $\text{SO}_2$ . The relative ratios of  $\text{NO}_x$  and  $\text{SO}_2$  removal can be adjusted to site specific needs by varying the concentration of the urea solution used in the hydration process. For a flue

gas containing 2500 ppm of  $\text{SO}_2$  and 750 ppm of  $\text{NO}_x$ , the lime-urea hydrate would have a molar ratio of urea to calcium of 0.075 to produce  $\text{Ca/S}=2$  and  $\text{urea/NO}_x=0.5$  when injected. This translates into a mass injection rate only 6 percent higher for the lime-urea hydrate than for  $\text{Ca(OH)}_2$ .

The lime-urea hydrate is effective in removing  $\text{SO}_2$  from combustion gases when injected into the upper furnace region of a boiler where gas temperatures are typically  $850^\circ\text{C}$  to  $1200^\circ\text{C}$ . The urea reactions take place over a temperature range of  $850^\circ\text{C}$  to  $1100^\circ\text{C}$  producing  $\text{N}_2$  and  $\text{H}_2\text{O}$ . After injection, the spent calcium portion of the sorbent can be captured and removed with standard precipitator or baghouse equipment. An additional advantage of the combined  $\text{NO}_x/\text{SO}_2$  process over selective non-catalytic  $\text{NO}_x$  reduction alone is the potential for eliminating, or markedly decreasing, air heater deposits from ammonium bisulfate and sulfate. These deposits are formed when residual ammonia reacts with  $\text{SO}_3$  formed during the combustion of sulfur bearing fuels. It is anticipated that the calcium will remove the  $\text{SO}_3$  at a relatively high temperature. Thus, it will not be available to react with any byproduct  $\text{NH}_3$  at air heater temperatures.

### OBJECTIVES

The specific objectives of the present study reported in this paper were to:

- o Assess the effect of urea addition to the water of hydration on the production rate and physical-chemical characteristics of calcitic quicklime.
- o Determine the levels of  $\text{NO}_x$  and  $\text{SO}_2$  removal attainable with the lime-urea hydrate.
- o Investigate any byproduct emissions of the  $\text{NO}_x/\text{SO}_2$  removal process, specifically  $\text{NH}_3$ .
- o Perform a preliminary economic assessment of the  $\text{SO}_2/\text{NO}_x$  removal process.

### TEST RESULTS

#### BATCH HYDRATION TESTS

A series of bench-scale hydrations were conducted using calcitic quicklime to determine the effect that the urea solution has on the hydration process, and the resulting properties of the hydrates. Of particular interest was the impact that urea has on the hydration time, since this could impact the size of the hydrator and capital cost.

Batch hydration experiments were performed using a stainless steel mixing container with manual stirring under a ventilated hood. Twice the stoichiometric amount of water was used for the hydrations. The temperature of the hydrate was monitored using a type K thermocouple with an analog output to a strip chart recorder. Samples of the hydrates were analyzed for C, H, N using a Perkin-Elmer elemental analyzer and particle size determined by sedimentation. The surface area of each hydrate was determined by single point BET analysis.

During these batch hydration tests, the material temperature was monitored on a strip chart recorder. The total hydration time was taken to be the time from the addition of the hydrating liquid to the point where the



material started to rapidly cool. The results of the time and temperature study are shown in Figure 2. The data for urea/CaO molar ratios of up to 0.2 (which is the highest anticipated for the majority of applications) indicate there was no effect on the hydration time. The hydration time subsequently increased as the urea concentration approached saturation.

To determine the effect of urea on the chemical properties of the hydrate, each hydrate was analyzed for C, H, N, and surface area. The results from the chemical analysis showed that the base quicklime contained a very small amount of carbon and hydrogen. The nitrogen values measured in the urea hydrates indicate that all of the urea was incorporated into the hydrate and was not lost as the excess water vaporized.

The surface area results are plotted in Figure 3. As seen in Figure 3, the surface area of the finished hydrate decreased with increasing amounts of urea. However, previous investigations (1, 2) have shown that sorbent utilization depends on in-furnace, not sorbent feed, surface area. Hence, the true measure of potential urea impact on the hydrate is the sulfur capture performance of the mixture compared to the standard hydrate.

Particle size distributions were determined by sedimentation for both calcium hydroxide without urea and the baseline urea-hydrate (.075 urea/CaO molar). A urea/CaO molar ratio of 0.075 corresponds to a Ca/S ratio of 2 and N/NO<sub>x</sub> ratio of 1 for flue gas containing SO<sub>2</sub> and NO<sub>x</sub> levels of 2500 ppm and 750 ppm, respectively. The mass mean diameters (MMD) of the hydrates with and without urea differed by less than 1 micron (MMD=3.5 microns without urea; MMD=2.7 microns with urea). Thus, at the urea levels of practical interest, the urea does not affect the particle size of the hydrate.

## SO<sub>2</sub> AND NO<sub>x</sub> REMOVAL CHARACTERISTICS

### PILOT-SCALE COMBUSTOR

Testing of the lime-urea hydrate was carried out in a pilot-scale combustor which fires natural gas doped with SO<sub>2</sub> and ammonia. The ammonia added at the burner is oxidized to NO<sub>x</sub> allowing control of the initial NO<sub>x</sub> level. This facility has been used for dry sorbent SO<sub>2</sub> removal studies in the past and is described in detail elsewhere (9,10). A diagram of the combustor is shown in Figure 4. The unit consists of three main sections: combustor section, injection section, and test section. Natural gas is burned in the combustor section and doped with SO<sub>2</sub> and NH<sub>3</sub>. Movable heat exchangers are used in the combustor section to control temperatures. Exiting the combustor section, the combustion products enter the sorbent injection section, comprised of a venturi, where the sorbent is injected through two diametrically opposed injectors. The gas sorbent mixture then flows through the 15 cm diameter, 241 cm long test section. Samples for either gas or solids analysis can be obtained at 15 cm increments along the test section using a water-cooled gas quench probe. For the data reported in this paper, gas samples were obtained at the exit of the combustor, and SO<sub>2</sub> and NO<sub>x</sub> removals determined using continuous gas analyzers. Byproduct ammonia was measured at the combustor exit using a water-cooled probe with a quartz wool filter. Two cubic feet of combustion products were passed through an impinger train containing dilute sulfuric acid. The impinger solutions were then analyzed for NH<sub>3</sub> with a specific ion electrode.

For the data reported here, the injection temperature is defined as the temperature measured at port S1, 28 cm downstream of the point of sorbent

injection, after the sorbent transport air and main combustion products are thoroughly mixed. This temperature is taken to be representative of the injection temperature, rather than the temperature just upstream of the point of sorbent injection, due to the amount of transport air used in the experiment. The sorbent transport air comprises 15 to 20 percent of the main combustion products in order to insure rapid and complete mixing.

Tests were performed at 750 ppm  $\text{NO}_x$  and 2500 ppm  $\text{SO}_2$  with the baseline lime-urea hydrate formulated to provide  $\text{N}/\text{NO}_x=1$  at  $\text{Ca}/\text{S}=2$ . Curves of  $\text{SO}_2$  removal versus  $\text{Ca}/\text{S}$  ratio and  $\text{NO}_x$  removal versus  $\text{N}/\text{NO}_x$  ratio were generated at 7 temperatures ranging from  $800^\circ\text{C}$  to  $1200^\circ\text{C}$ . On each day that the lime-urea hydrate was tested, a  $\Delta\text{SO}_2$  vs.  $\text{Ca}/\text{S}$  curve was also generated with a baseline or reference calcium hydroxide at the same running conditions. The tests with calcium hydroxide were used only to give an indication of any day-to-day fluctuations in the system performance so that any changes in performance due to the lime-urea hydrate could be properly evaluated. The data from these  $\text{Ca}/\text{S}$  and  $\text{N}/\text{NO}_x$  curves are plotted in Figure 5 for each temperature at  $\text{Ca}/\text{S}=2$  and  $\text{N}/\text{NO}_x=1$ , as well as  $\text{Ca}/\text{S}=3$  and  $\text{N}/\text{NO}_x=1.5$ . As seen in this figure, the optimum temperature for  $\text{NO}_x$  removal is approximately  $955^\circ\text{C}$  while the maximum  $\text{SO}_2$  removal is attained at about  $1100^\circ\text{C}$ . Further, the level of  $\text{SO}_2$  and  $\text{NO}_x$  also depends on the  $\text{Ca}/\text{S}$  and  $\text{N}/\text{NO}_x$  ratio. At the peak removal temperatures, the  $\text{NO}_x$  removal is 78 percent at  $\text{N}/\text{NO}_x=1.5$  and 53 percent at  $\text{N}/\text{NO}_x=1.0$ . The  $\text{SO}_2$  removals are 74 percent at  $\text{Ca}/\text{S}=3$  and 50 percent at  $\text{Ca}/\text{S}=2$ . Also shown in Figure 5 is the byproduct ammonia emitted at each temperature. As the temperature drops below the peak  $\text{NO}_x$  removal temperature,  $950^\circ\text{C}$ , the ammonia emission increases significantly and is also dependent on the  $\text{N}/\text{NO}_x$  ratio. As the injection temperature increases above  $955^\circ\text{C}$ , however, the ammonia emissions decrease significantly and are not dependent on the injected  $\text{N}/\text{NO}_x$  ratio.

Due to the increased ammonia emissions encountered at lower temperatures, injection temperatures of interest are between  $980^\circ\text{C}$  and  $1010^\circ\text{C}$ . The  $\text{SO}_2$  and  $\text{NO}_x$  removal performance of the lime-urea hydrate at  $1000^\circ\text{C}$  is shown in Figures 6a and 6b. Also shown in these figures is the performance of calcium hydroxide without urea, and a physical mixture of calcium hydroxide with pulverized urea. As seen in Figure 6a, the  $\text{SO}_2$  removal characteristics are not degraded by the presence of urea either in the lime-urea hydrate, or as a physical mixture of  $\text{Ca}(\text{OH})_2$  and urea. Figure 6b compares the  $\text{NO}_x$  removal performance of the lime-urea hydrate and the physical mixture of calcium hydroxide and urea. These data indicate that greater  $\text{NO}_x$  removal is attained when the urea is incorporated as a hydrate rather than injected as a physical mixture. At the present, reasons for the enhanced  $\text{NO}_x$  removal of the lime-urea hydrate over the physical mixture are not known.

Further pilot-scale combustion tests were conducted to determine if the urea to  $\text{CaO}$  ratio in the hydrate affected the  $\text{SO}_2$  and  $\text{NO}_x$  removal. Figures 7a and 7b show the  $\text{SO}_2$  and  $\text{NO}_x$  removal characteristics for three different lime-urea hydrates with urea/ $\text{CaO}$  molar ratio of 0.0375, 0.075 and 0.1125 at  $950^\circ\text{C}$ . Injecting these hydrates into a flue gas with 2500 ppm of  $\text{SO}_2$  and 750 ppm of  $\text{NO}_x$  at a rate yielding  $\text{Ca}/\text{S}=2$  corresponds to  $\text{N}/\text{NO}_x$  ratios of 0.5, 1.0 and 1.5, respectively. Conversely, the  $\text{N}/\text{NO}_x$  ratio would be 1.0 if the flue gas contained 375 ppm, 750 ppm or 1125 ppm  $\text{NO}_x$ , respectively. As seen in Figure 7, the urea/ $\text{CaO}$  ratio for the hydrate had no effect on the  $\text{SO}_2$  or  $\text{NO}_x$  removal characteristics. This indicates that the hydrate can be tailor made to site specific situations. Further pilot-scale tests are in progress investigating the effect of initial  $\text{SO}_2$  and  $\text{NO}_x$  ratios on the performance of the lime-urea hydrates.

## PROCESS ECONOMICS

An economic assessment of the process has just been initiated, and detailed results are not as yet available. However, a preliminary indication of the economics can be obtained from the following simplified analysis. Kaplan et al. (11) estimated the levelized cost of  $\text{SO}_2$  control from the dry furnace injection of  $\text{Ca}(\text{OH})_2$  to be approximately \$1000/ton  $\text{SO}_2$  removed. (Figure 8 outlines the basic assumptions used in the analysis by Kaplan et al.) An order of magnitude estimate of the cost for the lime-urea hydrate can be obtained by assuming: 1) no additional capital equipment is needed beyond that for dry  $\text{Ca}(\text{OH})_2$  injection; 2) urea is purchased at \$200/ton; and 3) in addition to the  $\text{SO}_2$  removal, the lime-urea hydrate results in 50 percent  $\text{NO}_x$  removal at  $\text{N}/\text{NO}_x=1.0$ . With these assumptions, the levelized cost of combined  $\text{SO}_2/\text{NO}_x$  removal is shown in Figure 8 as a function of the initial  $\text{NO}_x$  level. Note, in Figure 8, an initial  $\text{NO}_x$  level of zero corresponds to dry  $\text{SO}_2$  removal alone. As seen in Figure 8, the levelized cost-effectiveness of simultaneous  $\text{SO}_2/\text{NO}_x$  removal with the lime-urea hydrate can be 30 percent less than dry  $\text{SO}_2$  removal alone.

Again, a more detailed economic analysis is in progress. This ongoing work will both develop the capital and levelized costs of combined  $\text{NO}_x$  and  $\text{SO}_2$  removal with a lime urea hydrate, and compare the costs to alternate scenarios for  $\text{NO}_x$  and  $\text{SO}_2$  removal.

## CONCLUSIONS

The results to date indicate that the furnace injection of a single dry sorbent, lime-urea hydrate, results in simultaneous  $\text{SO}_2$  and  $\text{NO}_x$  removal. At an injection temperature of  $950^\circ\text{C}$ ,  $\text{SO}_2$  and  $\text{NO}_x$  removals of 50 percent are achievable with a lime-urea hydrate formulated and injected at a rate corresponding to  $\text{Ca}/\text{S}=2$  and  $\text{N}/\text{NO}_x=1$ . Larger  $\text{SO}_2$  and  $\text{NO}_x$  removals can be achieved by using higher sorbent injection rates. Or, if the lime-urea hydrate is used in combination with low- $\text{NO}_x$  burners (LNB), total  $\text{NO}_x$  reductions on the order of 75 percent are potentially achievable. This level of reduction is not only comparable to low- $\text{NO}_x$  burners in combination with selective catalytic reduction (SCR) (approximately 90 percent total  $\text{NO}_x$  reduction), but also more cost effective. Combining the  $\text{SO}_2$  and  $\text{NO}_x$  removal into a single sorbent eliminates the need for additional capital expenditures over that for dry sorbent injection alone, lowering levelized costs for  $\text{NO}_x$  and  $\text{SO}_2$  removal by up to 30 percent. This provides an attractive option for retrofit of older units with limited, or uncertain, remaining life.

The lime-urea hydrate process also has the potential advantage over other selective non-catalytic processes of minimizing air heater deposition by reactions between byproduct  $\text{NH}_3$  and  $\text{SO}_3$ .

Future work is planned to further develop this process. These activities will: 1) investigate ways to optimize the  $\text{SO}_2/\text{NO}_x$  removal process; 2) perform pilot-scale coal combustion tests; 3) investigate the potential beneficial impacts that the lime-urea hydrate will have on air heater deposition by ammonia/sulfur compounds; and 4) demonstrate the process at full scale.



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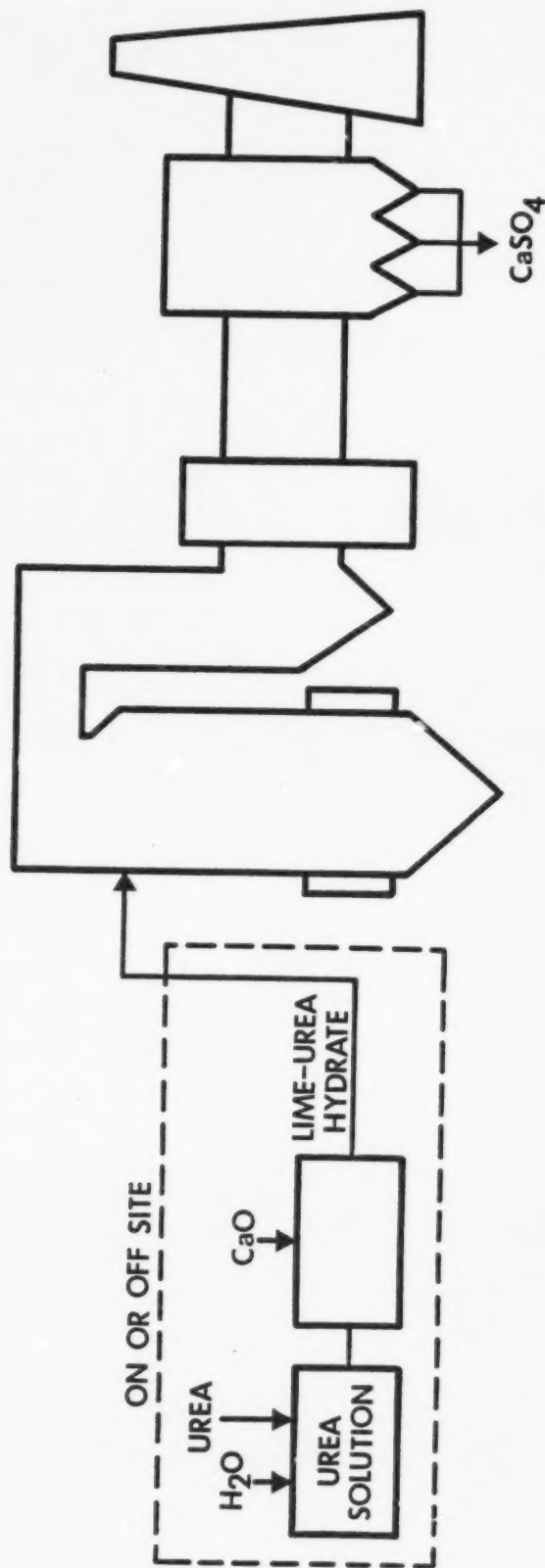


Figure 1. Simultaneous Dry  $\text{SO}_2/\text{NO}_x$  Removal Process Schematic

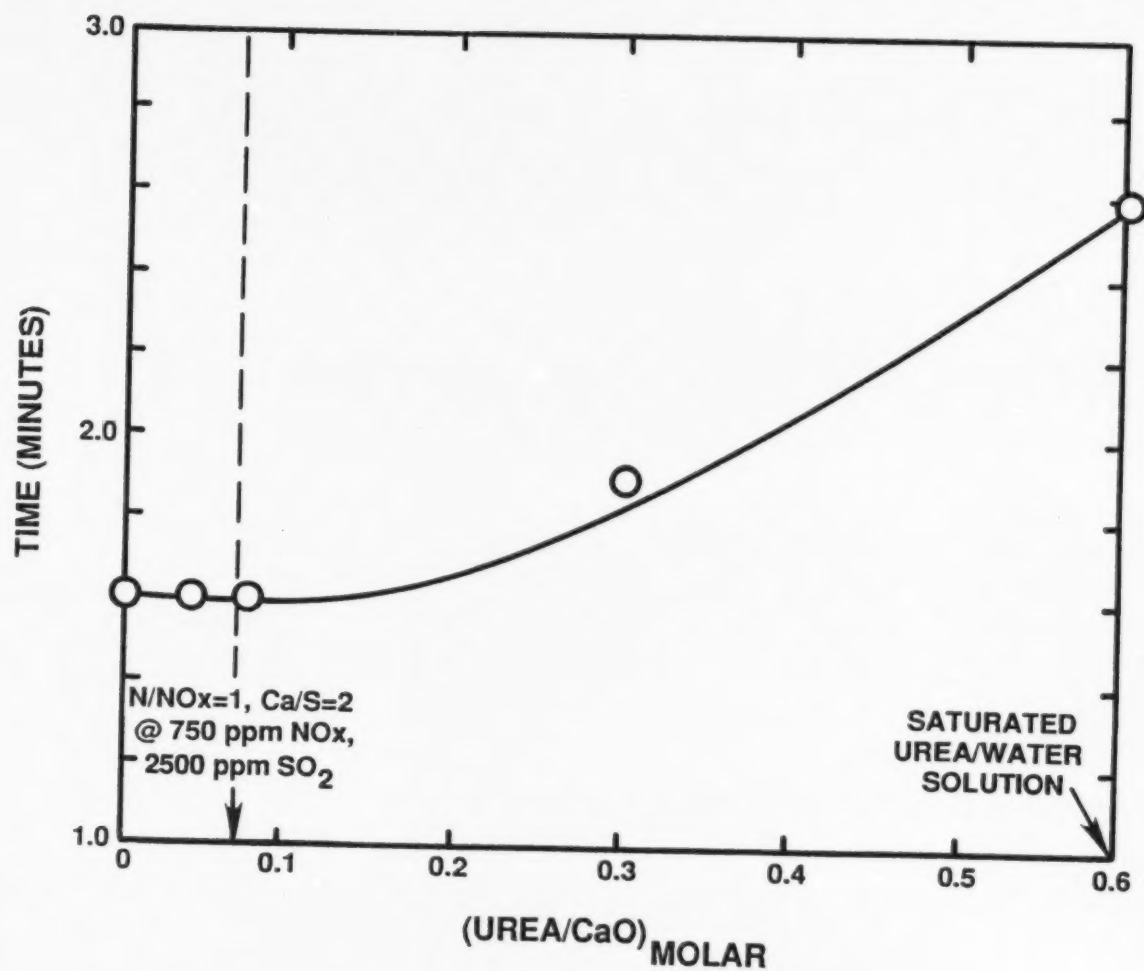


Figure 2. Effect of Urea on Hydration Time

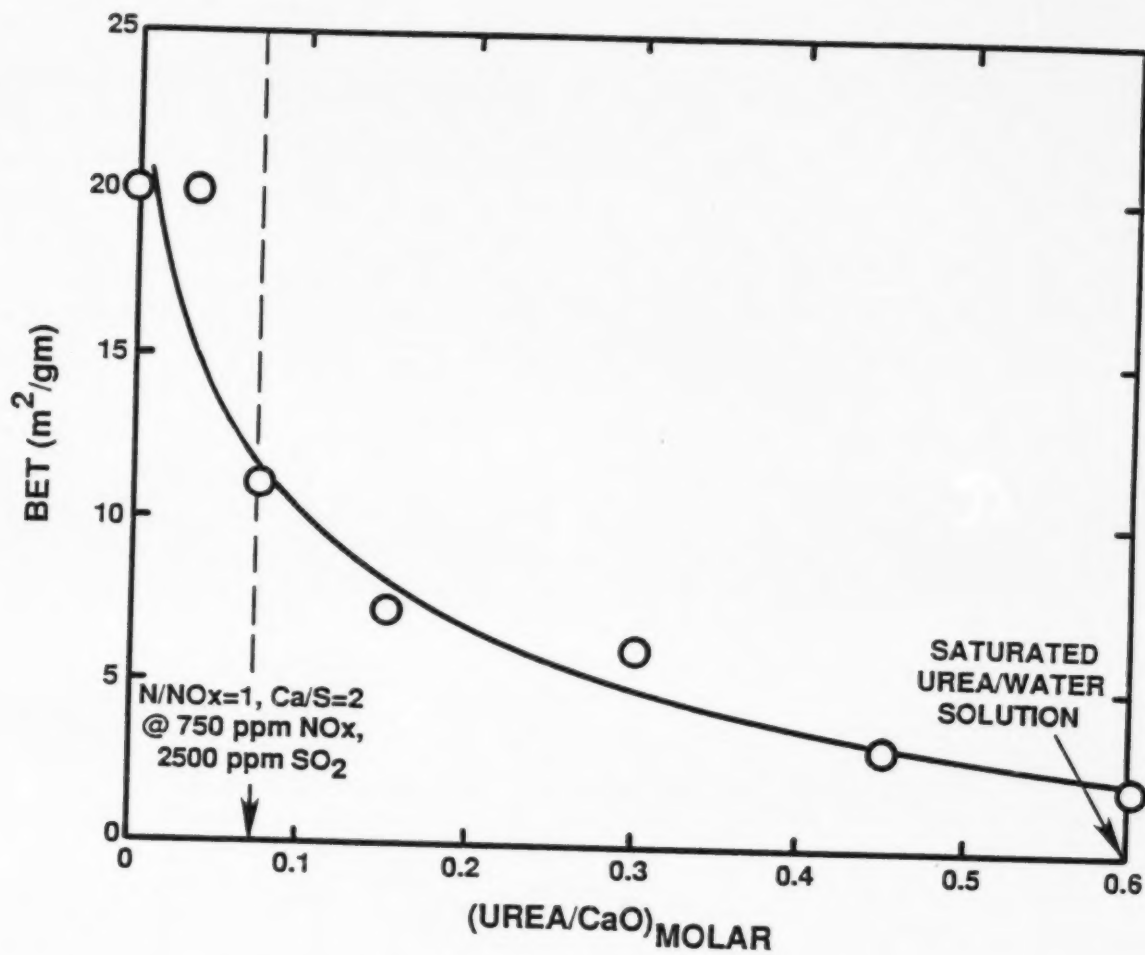


Figure 3. Effect of Urea on Hydrate Surface Area

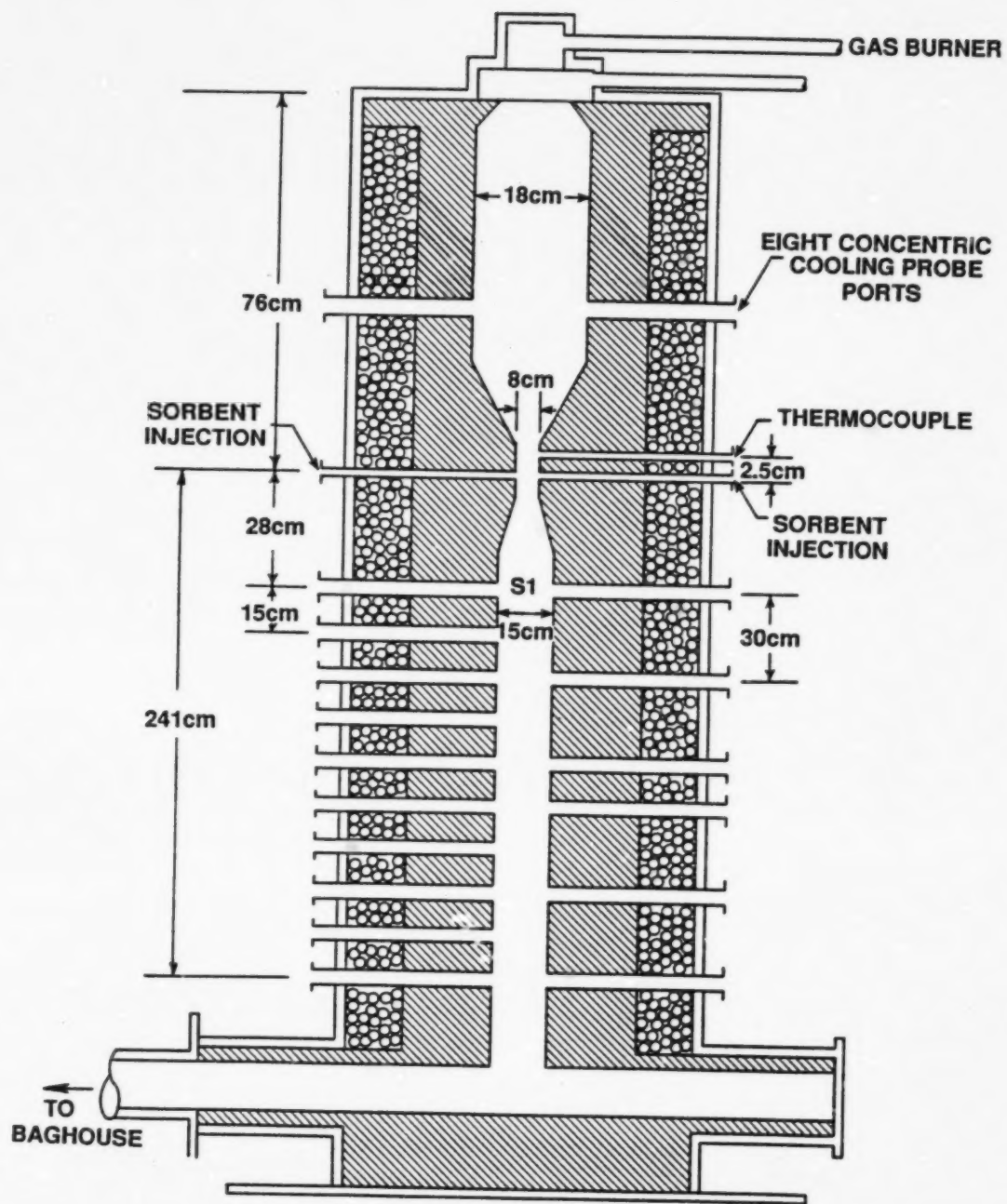
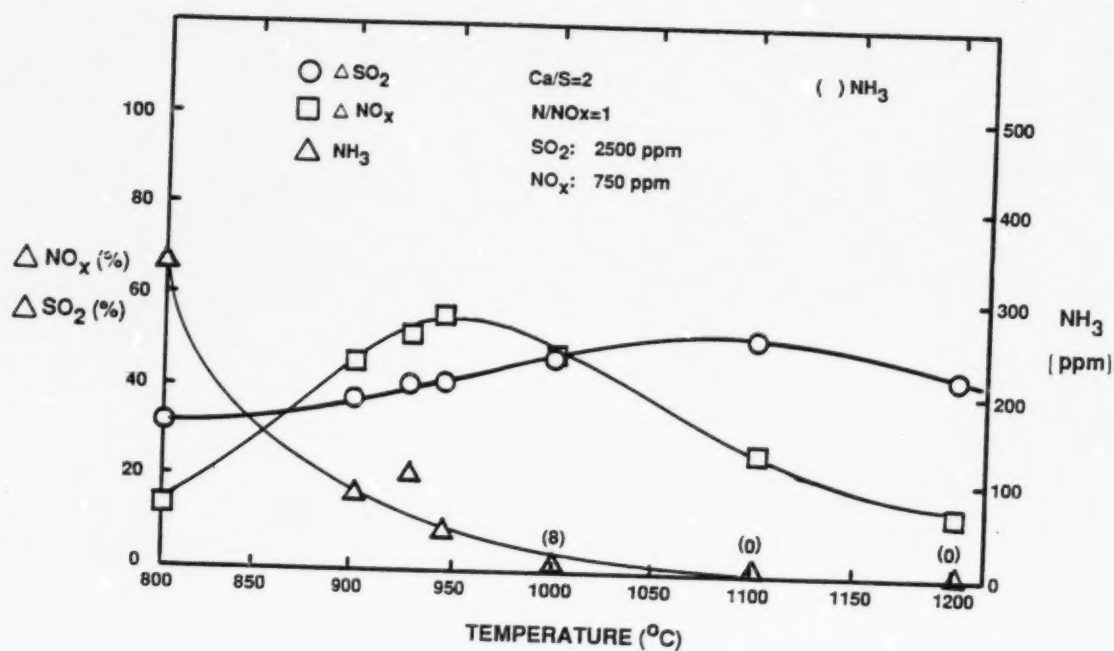
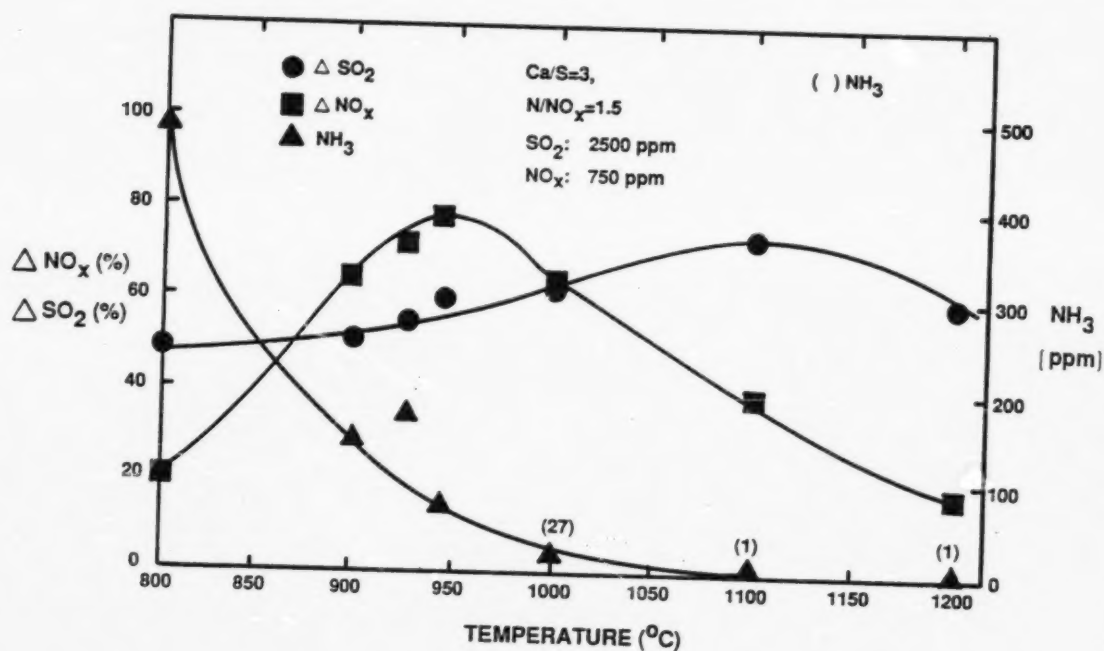


Figure 4. Pilot Combustor Schematic



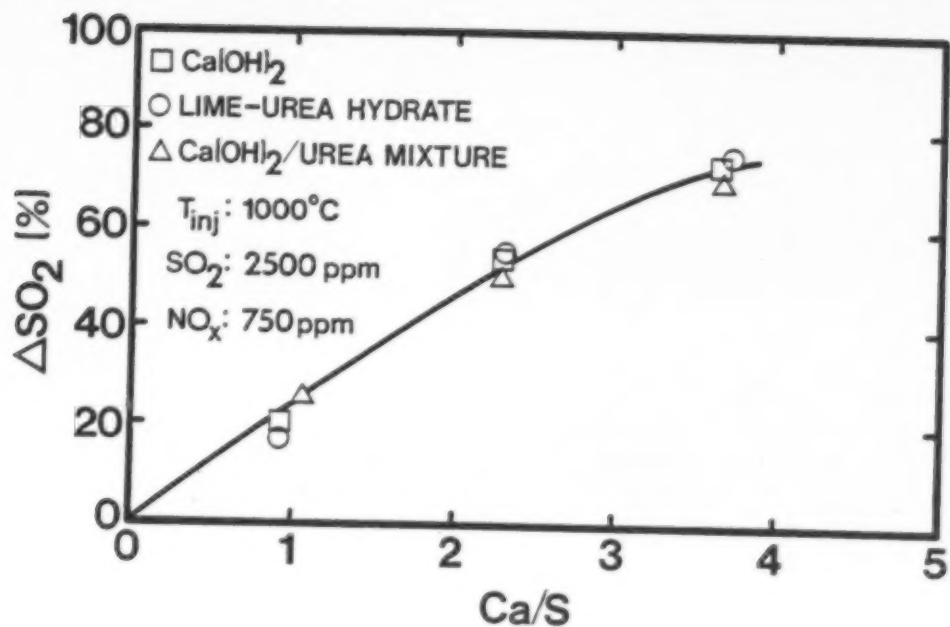


a. Ca/S = 2, N/NO<sub>x</sub> = 1

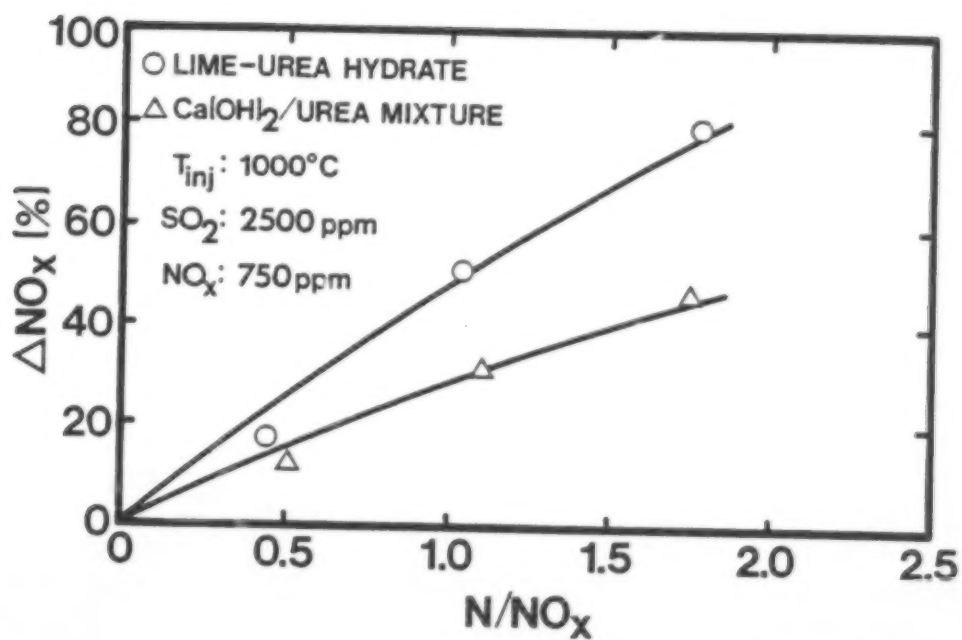


b. Ca/S = 3, N/NO<sub>x</sub> = 1.5

Figure 5. Effect of Temperature on the Performance of Lime-Urea Hydrate

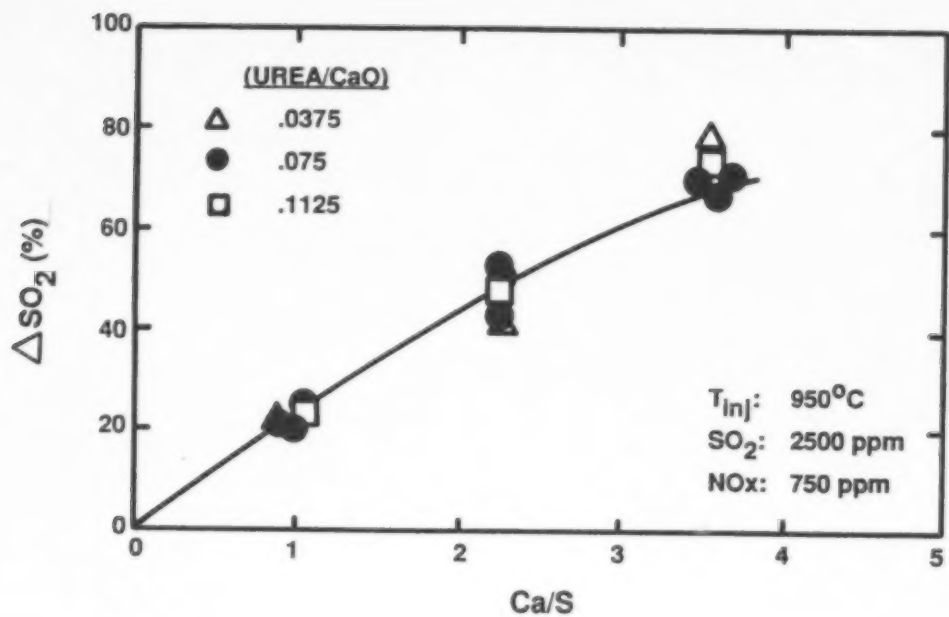


a.  $\text{SO}_2$  Removal

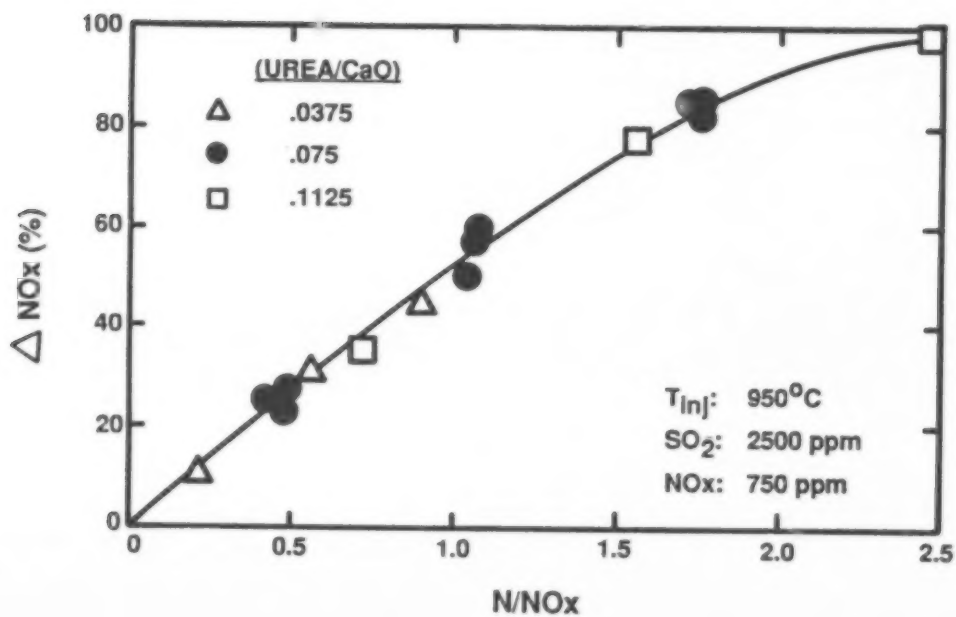


b.  $\text{NO}_x$  Removal

Figure 6.  $\text{SO}_2$  and  $\text{NO}_x$  Removal Characteristics of Sorbents with Urea



a.  $SO_2$  Removal



b.  $NO_x$  Removal

Figure 7. Effect of the Hydrate Urea/CaO Ratio on  $SO_2$  and  $NO_x$  Removal

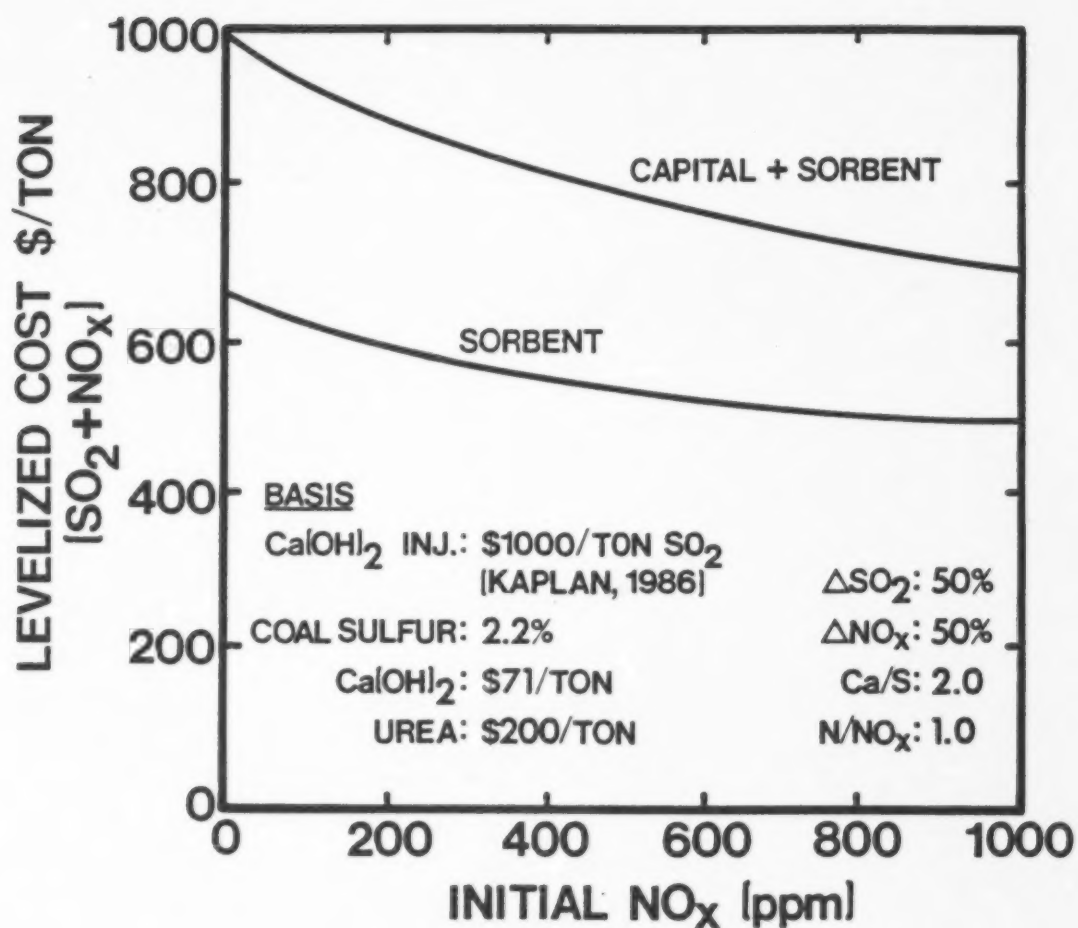


Figure 8. Preliminary Economics of Simultaneous Dry SO<sub>2</sub>/NO<sub>x</sub> Removal